## **OAK RIDGE** NATIONAL LABORATORY

MARTIN MARIETTA

## The Long-Term Problems of Contaminated Land: Sources, Impacts and Countermeasures

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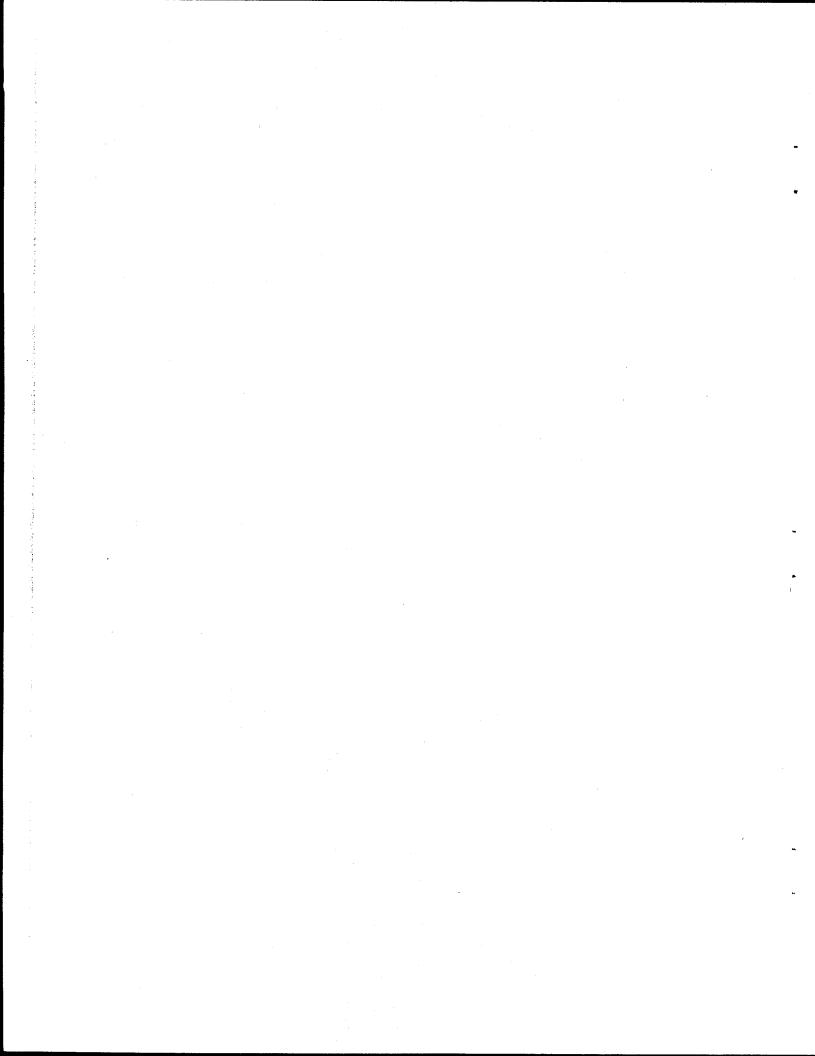
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#### **PREFACE**

This report is the first product of a project entitled "Residual Ecological Problems of Contaminated Land Areas." The project was supported by the Federal Emergency Management Agency (FEMA) under contract FEMA EMW-84-E-1737 with the Department of Energy (DOE 40-1457-84).

The work is intended as a state-of-the-art assessment of research in the subject area. Under no circumstances is this work intended as an original analysis of the subject area, but a review and synthesis of existing materials. The scope of the work includes ecological problems of contaminated land areas, including the potential effects of radiological contamination from a nuclear device, fallout radiation from nuclear attack, or contamination from hazardous waste or extensive dispersion of hazardous materials. However, because of the limited time available for this work and the enormous complexity in addressing the environmental behavior of hazardous materials, the majority of effort in this first report is in the area of land contamination by radionuclides. The second report issued under this project, ORNL-6239, title will deal exclusively with nonradioactive hazardous materials.

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#### SUMMARY

This report examines the various long-term environmental, health, and economic problems associated with contamination of land by radionuclides and hazardous chemicals. The areas addressed include both hypothetical and actual experiences with

- the extent of contamination;
- the impacts on man, agriculture, and the environment;
- countermeasures for mitigating exposures;
- radiological standards;
- alternatives for achieving land decontamination and cleanup; and
- possible alternatives for utilizing the land.

The report also identifies research needs. The body of information on radiological land contamination is extensive and impacts are well known. The literature on hazardous chemicals is also extensive, but impacts are more difficult to assess. Therefore, land contamination by hazardous chemicals is only briefly examined in this report by way of several example cases. A comprehensive treatment of the subject will be produced in a subsequent report, ORNL-6239.

Sources of land contamination by radionuclides considered in this report are nuclear war, detonation of a single nuclear device (e.g., a terrorist act), serious reactor accidents, and nuclear weapons accidents (termed "broken arrows"). Surface detonation of a single nuclear device or multiple detonations (nuclear war) would result in regional or worldwide contamination of the land with radioactivity. Radionuclides of concern from the standpoint of long-term land

contamination would be strontium-90, cesium-137, iodine-129, and the transuranics (plutonium, curium, and americium radionuclides). After surface detonation of the nuclear device, approximately 60% of the released radioactivity would be deposited within 24 h. After one year, approximately 5% of this deposited radioactivity (but only  $10^{-5}$  of the 1-h dose rate) would remain because of the decay of the short-lived radionuclides. Furthermore, after the first year, only about 10% of the radioactively contaminated land would give a lifetime dose greater than 10 rads to an unshielded person. (A rad is a unit of absorbed energy from ionizing radiation per unit mass equal to 0.01 J/kg). Ten rads is approximately the lifetime dose a person receives from natural background radiation (cosmic rays and natural radioactivity).

The most serious reactor accidents are characterized by a core meltdown, steam explosion, loss of containment, and dispersal of the nuclear fuels into the environment. However, such serious accidents are associated with low probabilities of occurrence (10<sup>-6</sup> to 10<sup>-7</sup> per reactor year). The extent of land contamination from a serious reactor accident would be regional, or roughly the same size area contaminated by the surface detonation of a one-megaton nuclear weapon. Like nuclear weapons contamination, strontium-90, cesium-137, and the transuranics would be the most important radionuclides of concern.

Hypothetical reactor accidents are much worse than any accident experienced so far. The worst reactor accident to date was the Three Mile Island (TMI) incident. In this accident, while some melting of the fuel did occur, there was no loss of containment, and only

radioactive isotopes of noble gases were released (therefore, no land contamination). Maximum radiation dose to an individual from TMI was predicted to be about 70 mrems ("rem" is a unit that quantifies the dose equivalent, which expresses all radiations on a common scale by multiplying the absorbed dose, in rads, by certain modifying factors that take into account the effectiveness and distribution of the absorbed dose). By comparison, the federal standard for nonoccupational doses is currently 500 millirems/year. The predicted average dose to the general public from TMI was roughly 0.8% of annual background dose.

Accidents involving nuclear weapons, or "broken arrows," result from mid-air collisions or crashes of aircraft carrying nuclear weapons, explosions of missiles carrying nuclear warheads, fires, and other such incidents involving nuclear devices. Since 1950, roughly 30 broken arrows have been verified, and there may have been as many as 200. A broken arrow does not include a fission event because of a number of safety design features built into nuclear devices to prevent accidental detonations. However, the arming mechanisms do involve conventional explosives, and dispersal of kilogram quantities of uranium and plutonium over square meter to square kilometer areas has occurred. Thus, some property losses have been associated with broken arrows, but no health effects from radiological contamination have been experienced.

The effects of radiation on man include radiation sickness and death at doses exceeding 100 rems over a short period of time. Also, there is a potential for increased cancer risk and genetic disorders

at lower doses. For example, it has been estimated by a number of health effects study groups that as many as 200 cancers would be induced in a population of a million exposed to the federal nonoccupational limit of 500 millirems/year. These 200 radiation-induced cancers would be a small percentage of the 250,000 "background" cancers naturally occurring in the population. Also, these low radiation dose risk estimates have been extrapolated from cancer risks at higher dose levels. The induction of cancers or genetic disorders from low doses of radiation has never been observed.

External doses would dominate after a nuclear war or a serious reactor accident. For example, one study predicted that 99.8% of the cancer deaths following a nuclear war would be from external exposures. Immediately after the contamination, external doses would be due primarily to cesium-134, cesium-137, ruthenium-106, cerium-144, and cobalt-60. After a year, approximately 95% of the initially deposited radioactivity (curies) would have decayed away, and after five years, external doses would be mainly from cesium-137. Weathering would reduce initial doses by half in the first two years. Further reductions are effected through the roughness of the ground surface and through shielding by structures.

Internal doses from extensive dispersion of radioactivity following a nuclear war or serious reactor accident would be much less important than external doses. In general, immersion in contaminated air or inhalation exposures are not as important as exposures from contaminated surfaces because resuspension accounts for only  $10^{-5}$  to  $10^{-9}$  of the initial surface activity. However, inhalation doses from the resuspension of plutonium would be important.

Ingestion exposures are generally not as important as external exposures because food-chain bioaccumulation involves transport across biological membranes, where the potential exists for discrimination against transport. For example, in the unlikely event that all of a person's food were grown near the detonation site, where the external dose rate from deposited strontium-90 would be equivalent to a gamma exposure rate of 100 roentgens (R)/h (1 R = 2.58 x 10<sup>-4</sup> C/kg of air) or greater, the annual ingestion dose from consumption of the food would be approximately 20 millirems/year. This is less than the Environmental Protection Agency (EPA) standard of 25 millirems/year for general public exposures from nuclear fuel cycle activities. Strontium-90 would account for most of this ingestion dose. Eighty to 100% of this dose would be from consumption of vegetables and produce. Less than 20% would be from consumption of milk and eggs, and less than 2% of the dose would be from beef, pork, and chicken meat consumption.

For contaminants other than radionuclides, it is also generally true that direct modes of exposure are much more important than indirect exposure routes, such as ingestion of contaminated food.

However, the food chain can be important if shortcuts bypass biological discrimination against transport. For example, soil ingestion by cattle bypasses the soil-plant uptake pathway. Also, if the chemical properties of the contaminant promote bioconcentration, the food chain can be important. For example, methyl mercury is more readily incorporated into the food chain than other (inorganic) forms of mercury. If the contaminant is very water soluble, and occurs in low-clay, low-organic soils, then root uptake may be quite high. Also,

if characteristics of the vegetation surface (e.g., leaf hairs, shape, and texture) promote adsorption or absorption of the contaminant, then ingestion exposures may be important. Finally, if the contaminant is persistent and lipophilic, then foods of animal origin may be important exposure routes.

The ecological consequences of a nuclear war might include:

- direct and indirect radiation effects on biota;
- the potential for increased biologically active ultraviolet (UV-B) solar radiation (290-320 nm wavelength) to reach the earth's surface from depletion of ozone in the stratosphere;
- microclimatic changes, such as soil erosion, temperature changes, and decreased humidity and rainfall in areas devastated by bomb blasts; and
- a global temperature decrease from suspension of dusts ("nuclear winter").

Direct and indirect radiation effects on natural systems might include loss of radiation-sensitive species and changes in vertebrate and insect populations from changes in food supply. Radiation sensitivity among plants is generally ordered conifers > hardwoods > shrubs > herbs > algae, lichens, and fungi. Such an order would lead to domination of "weedy" species in the aftermath of a nuclear war. However, recovery from radiation damage would be similar to that for fire, herbicides, and mechanical disturbances. Secondary succession (recovery) from severe devastation to a young pine forest requires 25-100 years.

Countermeasures for protection of livestock from radiation exposures following land contamination include sheltering (shielding) and placing them on stored feeds and protected water. If contaminated

feeds must be used, their intake could be reduced, or livestock could be fasted for short periods. Grains and concentrates would be preferred over pasture and hay to take advantage of the general ordering of bioaccumulation of contaminants in plants: fine roots > stems and leaves > seeds, fruits, and tubers.

If livestock consume contaminated feed, several measures can be taken to protect the consumer. Milk and eggs will be fairly safe for consumption after one week of feeding the animal uncontaminated feed. Clearance times for milk and eggs are quite rapid. Liver, kidney, internal organs, and bone meal should be avoided because these may be sites of contaminant deposition in the body, and their clearance times are relatively slow. On the other hand, muscle is relatively safe to consume. For iodine-131 contaminated milk, storage or diversion to manufactured products is the key. Iodine-131 has an 8-day radiological half-life (time it takes for the radioactive substance to lose 50% of its activity).

For vegetables and produce grown in contaminated soils, protection against contamination can be afforded through modifications to normal agricultural procedures or through avoidance of certain plant parts. In the aftermath of a nuclear war it would be prudent to plow under crops directly exposed to early fallout. However, contamination of subsequent crops could be minimized by deep plowing the soil to turn under surface contamination and mix it with uncontaminated soil. Also, the use of deep-rooted crops is recommended. The addition of calcium (lime) to soil generally reduces strontium uptake by plants, and

addition of potassium (potash) generally reduces cesium uptake.

Organic matter will also tend to bind a wide variety of contaminants,
making them less available for plant uptake from soil. The
substitution of "fruit" crops for "leafy" crops takes advantage of
the generally lower uptake of contaminants into reproductive plant
parts, and switching to fiber-producing crops avoids the food chain
altogether. If crops exposed to contamination must be consumed, then
outer leaves, peels, and shells should be avoided. Washing and
processing can remove much contamination. Also, cooking can thermally
destroy many organic contaminants.

There are several federal radiological standards which might apply in the event of land contamination following detonation of a nuclear weapon or a serious reactor accident. The Nuclear Regulatory Commission (NRC) has established a 5 rems per year occupational limit for workers in the nuclear industry (10 CFR 20). The nonoccupational limit is 500 millirems/year. The EPA has established a 25 millirems/year limit for doses to the public from nuclear fuel cycle activities (40 CFR 190). For uranium mining and milling activities. the EPA has set a standard of 15 pCi/g Ra-226 (a picocurie is a unit of activity equal to 2.22 disintegrations per minute), averaged over the top 15 cm of soil, for cleanup of mill tailings or debris from uranium processing sites (40 CFR 192). A very robust "standard" adopted by the NRC in licensing nuclear facilities is to keep emissions "as low as reasonably achievable" (10 CFR 50). It is difficult to predict which (if any) standards will apply in a given situation. However, standards should serve only as guidelines. In the aftermath of a nuclear war,

existing standards would undoubtably have to be revised upwards, relaxed, or ignored.

Methods of decontamination for man-made structures include firehosing, sandblasting, fixing, and asphalting contaminated surfaces. Reclamation of contaminated lands involves scraping, grading, vacuuming, or bulldozing and removing topsoil for treatment or disposal. In many cases contaminated soil, vegetation, and man-made structures are removed and buried or incinerated. If cleanup is not a viable option, then contaminated areas are usually fenced off to prevent unauthorized access.

There are several natural mechanisms which remove and disperse contamination. Leaching, surface runoff, and soil erosion following precipitation are natural dispersive mechanisms which must be addressed in the management of contaminated lands. Other dispersive mechanisms include resuspension (wind blown dusts) and animal vectors (burrowing and grazing animals). Radioactive decay and environmental degradation are removal mechanisms which aid in the cleanup of contaminated land.

Control of surface runoff can be achieved through construction of drainage ditches, dikes, dams, culverts, and diversion canals.

Infiltration of precipitation (leaching) can be controlled through land stabilization and use of surface seals and clay caps. Monitoring is necessary to determine effectiveness of cleanup operations and requires analytical support. For organic contaminants, there may be ways to treat and detoxify contaminated soils. Security will be required to limit access (exposures) by the general public. Also, records will need to be kept on cleanup procedures, monitoring results, and any

treatments to contaminated areas. Finally, a risk assessment should be performed to provide a perspective of the problem, determine cleanup levels, and estimate health and environmental impacts of the contamination.

Based on experience with the Enewetak (formerly "Eniwetok") Atoll cleanup, priorities for cleanup should be ordered in the following manner: (1) areas of human occupation, (2) intensively managed agricultural lands, (3) unmanaged agricultural lands, and (4) natural and recreational areas. Once these priorities have been established, the question of what level of cleanup must be achieved must be addressed. There are no universal rules. Each case must be individually addressed. Certainly if applicable federal standards are met, then no further cleanup may be necessary. However, since federal standards may not be applicable, other criteria may need to be adopted. For example, when the effectiveness of cleanup diminishes as the cost for further cleanup increases, then a practical limit may have been reached. Also, if the health and environmental impacts of the cleanup itself exceed the impacts from the land contamination, then a practical limit to cleanup may also have been reached.

In general, costs for cleanup of contaminated land are very high. The cleanup of the broken arrow in Palomares, Spain, was approximately \$33,600/ha (\$83,000/acre). Cleanup of the dioxin contamination in Seveso, Italy was approximately \$64,800/ha (\$160,000/acre). The purchase of evacuated residences at Love Canal, New York was \$7 million alone. The estimated cost for proposed cleanup of PCBs in the

Hudson River is \$10 million. Cleanup of Kepone in the James River is estimated at \$3 billion. Additional estimated cleanup costs are:

- \$2.20/kg (\$1/lb) of soluble substance on land
- \$1.85 per liter (\$7/gal) of floating substance on water
- \$53 to \$320 per liter (\$200 to \$1,200/gal) of insoluble substance on land or sinking substance on water,
- \$160 per liter (\$600/gal) for PCB spills
- \$860 to \$8,200/ha (\$350 to \$3,300/acre) for fencing off land
- \$1,200 to \$7,400/ha (\$500 to \$3,000/acre) for land stabilization with vegetation
- 2,200 to \$12,000/ha (\$900 to \$5,000/acre) for deep plowing
- \$5,900 to \$35,000/ha (\$2,400 to \$14,000/acre) for a soil cover or cap
- \$4000 to \$25,000/ha (\$1,600 to \$10,000 per acre) for soil removal and onsite disposal
- \$270,000 to \$1,500,000/ha (\$110,000 to \$600,000/acre) for soil removal and off-site disposal.

If contaminated land cannot be cleaned up, there are some possible alternative uses to which the land may be put. Production of fiber crops, seed-stock crops, or plant biomass for alcohol fuels are alternatives which divert agricultural land from food production. Conversion of cropland to pasture or rangeland takes benefit from the generally lower bioaccumulation of contaminants into animal products. Also, pasture and rangeland require little active land management. Finally, contaminated land areas could be fenced and used for the production of timber, either through the action of natural secondary forest succession or through active aerial seeding. Such an

alternative allows for 25-100 years or more of radiological decay or environmental degradation of contaminants before recovery of the timber.

For assessment of the health and environmental impacts from and the management of land contaminated by radionuclides, information needs are available in a large body of literature. Radiation effects on man, livestock, and vegetation and countermeasures for control of exposures are well documented. However, this work identified many information needs for land contamination by hazardous organic chemicals. There is a need for persistence data, toxicology information, quality control methods, and analytical procedures for hazardous organic chemicals. Also, there is a need for improved models for organics. Predictions of environmental transport and fate of hazardous organics are associated with a great deal of uncertainty.

#### 1. INTRODUCTION

This work is sponsored by the Federal Emergency Management Agency (FEMA) which is responsible for the management or coordination of a broad range of emergency management programs, including Federal Preparedness, Civil Defense, Earthquake Preparedness, Floodplain Management, Radiological Emergency Preparedness, and Disaster Assistance. FEMA has requested Oak Ridge National Laboratory (ORNL) to undertake a series of "state-of-the-art assessments of research" on a number of topics, including (1) geophysical consequences of a nuclear exchange; (2) crisis evacuation; (3) emergency shelter, food, and water; (4) postdisaster economic recovery; and (5) contaminated land areas. The main purpose of these assessments is to provide FEMA with a review and synthesis of existing information related to these subjects so that officials will be more knowledgeable on subject matters bearing on their decisions. Also, the intent of the work is to identify areas where additional research may be needed.

The material in this report addresses residual ecological problems of land areas contaminated by radionuclides and hazardous chemicals, but the report focuses primarily on the extent, impacts, and cleanup of contamination from major reactor accidents, nuclear weapons fallout (nuclear war and single detonations), and nuclear weapons accidents not involving fission ("broken arrows"). The review considers impacts of radionuclides remaining in the soil at one year following initial deposition, and identifies potential effects on human health and long-term land use. Land contamination by hazardous chemicals is

briefly examined, but will be reviewed more extensively in a subsequent report, ORNL-6239.

In researching the literature on land contamination, nearly 100 documents prepared in the late 1950s and early 1960s for the U.S. Atomic Energy Commission were identified as potential information sources. Most of these documents are concerned with the effects of ionizing radiation on livestock and crops, radiation shielding, and decontamination. While this original work provides a basis for the review, additional, more recent compilations and discussions of this material have become available. Also, in the time since those early reports were published, the cleanup of the Enewetak (formerly "Eniwetok") Atoll Islands occurred (DOE 1982). This cleanup not only provided a wealth of experience with contaminated land, but the logistic and political problems that were addressed also served to establish a rationale for balancing the costs and benefits of land decontamination.

In early 1944 the U.S. Navy attacked Japanese forces occupying the Marshall (Enewetak Atoll) Islands and established a Navy and Marine Corp air base and fleet anchorage. Immediately after World War II, the U.S. began testing nuclear weapons on islands of the atoll.

Approximately 145 inhabitants of the islands were evacuated, and from 1946 to 1958, 43 nuclear devices were detonated. The nuclear weapons testing created radiological contamination of soils, water, and food that precluded unrestricted use of the islands by the returning Marshallese without prior cleanup. Thus, a cleanup program was established between 1972 and 1979 which had as its goal (DOE 1982)

"the removal of vegetative overgrowth, debris, and structures or materials ... which could pose radiation or other hazards to inhabitants, interfere with their reasonable use of the atoll, or preclude safe, continuous habitation."

Initial cleanup guidelines were established in 1972-1973 to determine the magnitude of the problem. For plutonium these guidelines included (DOE 1982):

- a cleanup to 500 pCi (a picocurie is a special unit of activity equal to 2.22 disintegrations per minute) residual Pu-239 per gram of soil in the top 5 cm of soil by either removing soil to a depth of 61 cm (24 in.) or covering with soil having negligible radioactivity and
- plowing to a depth of 25.4 cm (10 in.) any soil with surface contamination exceeding 50 pCi/g.

These priorities were intended as guidelines for determining scope-of-effort cost estimates. There was, however, much discussion of these guidelines in light of the proposed 1 mrad/year Environmental Protection Agency (EPA) Federal Guidance Standard (EPA 1977) and the tremendous costs involved. (A rad is a unit of absorbed energy from ionizing radiation per unit mass equal to 0.01 J/kg.) An advisory group was established to review the cleanup criteria, and they found that it was impossible to assure that no individual would be exposed to radiation levels above the EPA standard. Instead, they recommended criteria that would result, on the average, in radiation doses commensurate with the EPA standard. The actual cleanup was based on criteria established in 1978. These final criteria were:

- on food-gathering islands, cleanup all surface soil in areas greater than 0.5 ha where soil radioactivity levels exceed 160 pCi/g;
- on agricultural islands, cleanup all surface soil in areas greater than 0.5 ha where soil radioactivity levels exceed 80 pCi/g;
- on village islands cleanup all surface soil in areas greater than 0.25 ha where soil radioactivity levels exceed 40 pCi/g.

These final criteria, although specific for the Enewetak Islands and their particular contamination problems, are useful because in a broader sense, they established a rationale for cleanup and reclamation of contaminated land based on the potential for human exposures. The approach is to require most stringent controls on lands used for human occupation, more latitude on agricultural lands, and even more latitude on lands not intensively managed by man. This approach reflects the tradeoffs between potential health and safety issues (and uncertainties associated with estimation of health effects) and the costs and logistics of cleanup. The approach must be administered according to the specifics of the problem, and additional criteria may need to be established in specific cases. For example, considerably revised standards may be required in the aftermath of a nuclear war.

This report will review the accidents and events which can result in land contamination, the potential effects of various contaminants on human health and safety, and ways that remedial action can mitigate problems.

#### 2. SOURCES OF CONTAMINATION

#### 2.1 FALLOUT FROM NUCLEAR WEAPONS DETONATION

Large areas of land may be contaminated by radioactive fallout from nuclear weapons detonations. The extent of residual contamination ranges from regional, in the case of a single, low-yield detonation to worldwide in the case of a nuclear war. Approximately 200 radionuclides of 35 different elements have been identified from nuclear explosions and fallout. The radiological decay rate of these radionuclides is such, however, that after one year only about 5% of the total radioactivity, (curies) represented by a few long-lived radionuclides, remains. After one year the instantaneous dose rate is 10<sup>-5</sup> of the dose instantaneous rate at 1 h (Glasstone 1962).

The kind and amount of fallout available for contamination of land areas is a function of the type of nuclear device (fission or fusion weapon), the explosive yield (kiloton or megaton), and detonation point (surface or air burst). Barnaby and Rotblat (1982) review the early and late effects of nuclear weapons and, in consideration of the above, tabulate estimates of areas contaminated, exposures, dose rates, and accumulated doses.

#### 2.1.1 Source of Contamination

The major long-lived radionuclides produced by explosion of a one-megaton fission weapon are given in Table 2.1. These are the radionuclides that will be deposited over large areas of land if the device is detonated on, or very near, the earth's surface, and will be

Table 2.1. Major long-lived radionuclides produced by a one-megaton fission explosive  $\underline{a}$ 

Radionuclide Half-life <u>b</u> (years	Cur	ies produced
H-3 <u>c</u>	10,000,000	12.3
C-14	20,000	5,730
Co-60	300	5.3
Sr-90	88,000	28.6
Ru-106	2,100,000	1.0
I-129 <u>₫</u>	8,300	15,700,000
Cs-134	5,000	2.1
Cs-137	150,000	30.2
Ce-144	3,600,000	0.8
Pu-238 <u>d</u>	20	87.8
Pu-239 <u>d</u>	1,000	24,131
Am-241 <u>d</u>	400	432.2

aSource is Ng and Tewes (1971).

 $<sup>\</sup>underline{b}$ Source is Kocher (1981).

 $<sup>\</sup>underline{\mathtt{c}} \mathtt{From}$  detonation of a one-megaton fusion device.

dsource is NAS (1975).

present in relatively large quantities at one year after detonation. Four of these radionuclides, cobalt-60, ruthenium-106, cesium-134, and cerium-144, have half-lives of five years or less and will decay in a few years, but the others will remain as a source of contamination for decades. (A radiological "half-life" is the time required for the radionuclide to lose 50% of its activity.)

A single explosion of a nuclear weapon, whether by accident or by an act of terrorism, could be expected to produce the radionuclides listed in Table 2.1 in quantities largely dependent upon the fission yield of the nuclear device. Currently, many nuclear weapons are combined fission-fusion types; therefore, no single weapon will produce the exact quantities of radionuclides listed here. With explosive capabilities ranging from a few kilotons to several megatons, the radionuclide quantities can vary by orders of magnitude for different weapons. All of them, however, leave varying areas of land contaminated with the long-lived radionuclides strontium-90, cesium-137, iodine-129, and transuranics. These radionuclides could expose man and other biota to radiation for many years.

#### 2.1.2 Extent of Contamination

Fallout particles range in size from less than 1  $\mu m$  to over 1 mm in diameter. The heaviest particles fall to earth very quickly in the immediate vicinity of the burst, while lighter particles stay in the air for a longer period and may be carried by the wind for very great distances before depositing on the ground. About 60% of the total radioactivity from a groundburst is deposited in early fallout or

fallout deposited within 24 h after an explosion (Barnaby and Rotblat 1982). For an air burst (in which the fireball does not contact the ground), the fission products circulate in the upper atmosphere and do not form early fallout.

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After detonation of a nuclear weapon, the distance traveled by fallout particles and the time of their deposition are dependent upon the speed and direction of the wind. Under steady wind conditions, with no change in wind direction with height (shear), the radioactivity will be distributed in a roughly cigar-shaped pattern. However, changes in wind speed and direction or scavenging of the fallout particles by rain or snow can give rise to small areas where very large amounts of radioactivity are deposited. Thus, the idealized cigar-shaped isodose patterns may be greatly altered (Barnaby and Rotblat 1982).

Nevertheless, the idealized pattern is useful in providing an estimate of the distribution of fallout and radiation dose for a weapon detonation. The total or accumulated dose received by a person is the product of the dose rate (rads per hour) and the time of exposure. Since dose rate rapidly decreases with time (as a function of radionuclide decay), most of the accumulated dose is delivered early after a nuclear explosion. Barnaby and Rotblat (1982) assumed a constant wind velocity with little directional shear and no precipitation and estimated the length and width of contaminated areas (fallout pattern) for given reference dose rates. Table 2.2 gives the results of their calculations for a one-megaton thermonuclear weapon

Table 2.2. Areas covered by given accumulated doses from fallout from a one-megaton weapon≜

umulated dose <sup>b</sup> (rads Total	Residual	Area
(1 h)	(1 y)	(km <sup>2</sup> )
1000	60	90
800	48	1,20
600	36	1,70
400	24	2,60
200	12	5,50
100	6	10,50
50	3	18,60
25	1.5	32,70
10	0.6	56,00

<u>A</u>A thermonuclear weapon with half of the yield due to fission (no correction for structural shielding, ground surface roughness, and weathering of deposited material). Source is Barnaby and Rotblat (1982).

 $\underline{b}$ Integrated from the time indicated until complete radiological decay of the long-lived radionuclides.

with half of the yield due to fission. It gives the areas within which reference total (1 h postdetonation) accumulated dose could reach the value in the first column of Table 2.2. Since about 94% of the accumulated dose will be delivered within one year, these areas could be expected to produce the residual doses in the second column of Table 2.2 from one year postdetonation until complete radiological decay of the long-lived radionuclides. About 10 rads is the dose which individuals may accumulate in a lifetime exposure to natural background radiation, so approximately 9% (11,900 km²) of the contaminated area could be expected to produce radiation exposures which are one to several times background after one year. These estimates assume no effects from structural shielding, ground surface roughness, and weathering of the deposited fallout. Such effects will reduce radiation exposures by varying amounts, perhaps by a factor of 10 on the average.

An estimate of the extent of ground contamination from strontium-90 is given in Table 2.3 for a one-megaton weapon. In addition, the radionuclides listed in Table 2.1 could be expected to occur in various proportions with ruthenium-106, cesium-137, and cerium-144 being deposited in greater quantities than strontium-90.

Numerous estimates have been made of the extent of fallout contamination after a nuclear war (Miller and LaRiviere 1966; National Research Council 1975; Chester and Chester 1976). Of course, such estimates are dependent upon assumptions made for weapon sizes, deployment, and total explosive power employed. Table 2.4, for example, gives estimates of the average deposition of long-lived

Table 2.3. Deposition of Sr-90 from fallout of a one-megaton weapon detonated at ground level $\underline{a}$ 

Deposition of Sr-90 (Ci/km <sup>2</sup> )	Area (km²)
>1.5 0.5 to 1.5	3,750 7,500
0.15 to 0.5	12,000

aSource is Bondietti (1982).

Table 2.4. Average deposition of particulate radionuclide one year after detonation of 10,000 megatons of nuclear explosives  $\underline{a}$ 

Radionuclide	Deposition (Ci/km $^2$ ) $^{\underline{b}}$		
Sr-90	1.9		
Ru-106	20.3		
I-129	0.3		
Cs-137	2.9		
Ce-144	40.3		
Pu-238	. 0.001		
Pu-239	0.04		
Am-241	0.02		

 $\underline{a}$ Source is National Academy of Sciences (1975a). Other radionuclides produced (e.g., Cs-134) were not given.

 $\underline{b}$ Average deposition in latitudes 20-50°N.

radionuclides in the United States at one year following a nuclear war in which 10,000 megatons of nuclear explosives were used. Regions on the scale of thousands of kilometers in the northern hemisphere could be contaminated two to three times more than the average numbers indicate. Smaller regions, tens to hundreds of kilometers in size, could be subjected to levels up to an order of magnitude higher.

We estimate that contamination levels given in Table 2.4 would lead to radiation doses on the order of 700-800 millirems/year to unshielded individuals living in such areas. Ground roughness and weathering would reduce these dose estimates.

Chester and Chester (1976) have examined the consequences of a nuclear war in which commercial nuclear power reactors are targeted to damage the nation's industrial base and produce augmented fallout (weapons plus reactor fuels). They concluded that the strategic value of such an attack was marginal, but there would be an approximate doubling of the strontium-90 contamination on the more contaminated grain-producing land compared to that from the weapons' fallout alone. This would lead to approximately double the cancer death rate due to radiation exposure in the postattack environment compared to the total peacetime rate.

#### 2.2 REACTOR ACCIDENTS

In terms of contaminating large areas of land, serious reactor accidents have the greatest potential of any nonweapon device (U.S. NRC 1975). Although such accidents are very improbable  $(10^{-6} \text{ to } 10^{-7} \text{ per reactor year})$ , estimated releases of

radioactivity from the most serious accidents are about as great as those for nuclear weapons in the one-megaton range. Table 2.5 gives releases of long-lived radionuclides for the most serious reactor accident analyzed in the U.S. Nuclear Regulatory Commission's (NRC) Reactor Safety Study (U.S. NRC 1975). It should be pointed out, however, that reactor accidents having much higher probabilities of occurrence are far less serious in terms of radioactivity released and extent of contamination.

#### 2.2.1 Source of Contamination

The most serious potential light-water reactor accident is characterized by a core meltdown followed by a steam explosion in the containment vessel. The probability of occurrence of such an accident has been estimated to be is  $10^{-4}$  to  $10^{-5}$  per reactor year (U.S. NRC 1975). Assuming failure of heat removal systems, there is a loss of containment due to rupture of the reactor vessel, and a large amount of radioactive material (gases and particulates) is released into the environment. Depending upon meteorological conditions (wind speed and direction, atmospheric stability, and presence of precipitation during the accident), a plume of radioactive materials leaves the reactor site and, as it travels in the prevailing wind, radionuclides are deposited on the ground along the path of the plume. The total deposition would be approximately the same under both dry and wet conditions, but the fallout distribution would be somewhat different. That is, precipitation during the accident can cause heavier deposition of radioactivity near the accident site, compared to levels deposited by dry deposition processes.

Table 2.5. Long-lived radionuclides released in the most serious light-water reactor accidenta

Radionuclide	Half-life <u>b</u>	Curies released
Co-60	5.3 y	116,000
Sr-90	28.6 y	185,000
Ru-106	1.0 y	10,000,000
Cs-134	2.1 y	3,000,000
Cs-137	30.2 y	1,880,000
Ce-144	284.3 d	255,000
Pu-238	87.8 y	171
Pu-239	24,131 y	63
Pu-240	6,569 y	63
Pu-241	14.4 y	10,200
Am-241	432.2 y	5.1
Cm-242	163.2 d	1,500
Cm-244	18.1 y	69

acore meltdown with steam explosion and loss of containment (probability is 9 x  $10^{-7}$  per reactor year). Source is U.S. NRC (1975).

 $\underline{b}$ Kocher (1981).

The radionuclide releases listed in Table 2.5 are for the most serious reactor accident with a probability of 9 x  $10^{-7}$  per reactor year. Accidents having probabilities of occurrence of  $10^{-4}$  to  $10^{-5}$  per reactor year might be expected to have releases which are orders of magnitude less. Estimated radionuclide releases for the most serious accidents considered in the Reactor Safety Study may be much greater than could actually occur. Experience at Three Mile Island and new studies on accident source terms indicate that radionuclide releases associated with meltdown accidents may be much smaller than those assumed here.

### 2.2.2 Extent of Contamination

The deposition of fission product radionuclides upon land surfaces represents the source of contamination following a serious reactor accident. As is the case with weapons' fallout, there is a rapid decay of short-lived fission products, and most of the radiation dose is delivered during the first year following the accident. Only those radionuclides listed in Table 2.5 will remain as a significant source of contamination after one year. The more rapid decay of ruthenium-106, cerium-144, cobalt-60, and the smaller releases of transuranic radionuclides (plutonium, americium, curium) leave strontium-90 and cesium-137 as dominant contaminants from a dose standpoint.

Table 2.6 gives estimates of levels of contamination at one year following the accidental release (over a 2.5-h duration) of the

Table 2.6. Deposited radionuclides at one year following release from the worst reactor accident  $\underline{\underline{a}}$ 

Radionuclide -	Contamination at various distances (Ci/km $^2$ ) $^{\underline{b}}$			
	900 m	10,000 m	60,000 m	100,000 m
Co-60	488	12.6	0.9	0.05
Sr-90	868	22.6	1.7	0.08
Ru-106	20,048	521.2	38.9	1.9
Cs-134	10,279	267.1	19.9	1.0
Cs-137	8,816	229.2	17.1	0.9
Ce-144	714	18.6	1.4	0.07
transuranics	51	1.3	0.1	0.005

<u>a</u>Release of radionuclide quantities are given in Table 2.5.

 $\underline{b}$ Estimates obtained using the computer code AIRDOS-EPA (Moore et al. 1979) assuming all radioactivity is released downwind in a 22.5° corridor.

radionuclides given in Table 2.5. These levels of contamination could be expected to have decreased by about one-third in the five years following the accident. At this time, cesium-134 and cesium-137 would account for 19 and 69%, respectively, of the contamination.

By contrast, a reactor accident involving a large pipe break and no core melt is more probable ( $4 \times 10^{-4}$  per reactor year). It is estimated that an accident of this nature would release only about 4.5 Ci of cesium-134, 1.8 Ci of cesium-137, and  $4 \times 10^{-5}$  Ci of strontium-90 (U.S. NRC 1975). Other short-lived radionuclides would be released, but only the relatively long-lived radionuclides would produce long-term contamination of the surrounding land. An accident of this size would result in contamination levels at 1000 m distance of about 23 mCi/km<sup>2</sup>. External dose rate at one year would be about 3 millirems/year.

## 2.2.3 Experience

Past reactor experience in the United States has included several serious accidents. However, none of these has been harmful to public health. With the exception of the Three Mile Island accident, the serious events involving releases of radioactivity have involved experimental or prototype reactors.

2.2.3.1 The Enrico Fermi Accident. The Enrico Fermi, a prototype for a commercial fast breeder reactor, had a serious accident on October 6, 1966. The reactor, located at Newport, Michigan (about 30 miles from Detroit), was being operated at about 7% of its designed operating power of 430 MW(t).

Two pieces of zirconium plate came loose from their location inside the reactor pressure vessel and partially blocked the sodium coolant flow into four of the reactor's 103 core fuel subassemblies. The reduction in coolant flow created a temperature rise sufficient to damage the fuel rods, and some of the fission product activity in the damaged fuel was released into the reactor building. No measurable amount of activity escaped the building. Alarms caused the reactor building to be automatically closed, power reduction was begun, and in 11 min the reactor was shut down.

The reactor was repaired, and in 1970 it was again operational.

This accident demonstrated a reliable and orderly operation of the reactor crew and the reactors' accident limitation systems.

2.2.3.2 The SL-1 Accident. The SL-1, located at the National Reactor Testing station at Idaho Falls, Idaho, was a prototype of a boiling water reactor intended for military use as a power and heat source. Its design operating power was relatively small, 3 MW(t). On January 3, 1961, three men who were doing maintenance on the shutdown reactor were killed. Since these three men were the only people present at the time and the recording instruments were not operating, the exact sequence of events leading to the accident is uncertain. It is probable that a nuclear excursion caused by manual withdrawal of a control rod resulted in the rapid formation of high-pressure steam which destroyed the reactor.

The building which housed the SL-1 was designed to provide shelter rather than containment. There were no airlocks or seals to contain fission products and the building was not designed to withstand large

internal pressures. About 20% of the core plate area, which contained 40% of the fission product inventory, was destroyed in the accident. Only 5 to 10% of the fission product inventory escaped from the reactor vessel and only 0.01% got outside the rudimentary reactor building. If the reactor had been in a pressurized, operating state, the fission product release would probably have been greater. No member of the public received a detectable radiation exposure from the accident.

2.2.3.3 The Three Mile Island Accident. On March 28, 1979, the United States experienced the worst accident in the history of commercial nuclear power generation. Although no serious contamination of off-site property occurred, and public radiation exposures were low, this accident resulted in an investigation by a Presidential Commission and new evaluations of reactor safety, emergency preparedness, and response plans at all levels of government.

The Three Mile Island facility is located at Goldsboro,
Pennsylvania, on an island in the Susquehanna River just south of
Harrisburg. The plant is a pressurized water reactor that can produce
880 MW of electric power. On the morning of March 28, 1979, the
accident apparently started when a turbine "trip" let off steam into
the outside atmosphere at 690,000 Pa (100 psi). Failure of a feedwater
pump in the secondary cooling system caused pressure to rise in the
primary cooling system. A pressure relief valve opened, but failed to
close properly, creating a leak in the cooling system. Events followed
that led to a general emergency at the plant. Operator errors and
equipment failures resulted in coolant levels in the pressure vessel
falling below the top of the core which caused serious overheating.

Additionally, a hydrogen bubble formed in the reactor and for five days uncertainty prevailed as regards possible consequences of the accident. Early fears that the core would melt down and expel large amounts of radioactivity changed to fears that the hydrogen found in the reactor might explode. General confusion among utility and federal and state officials about potential consequences and whether or how to evacuate surrounding populations pointed to the need for reassessment of emergency plans for evacuation following serious reactor accidents and for establishment of clear lines of communication and authority.

The reactor core was severely damaged, but did not completely melt down. The total release of radioactivity to the environment was established at 13 to 17 li of radioiodine and from 2.4 to 13 million curies of noble gases (Kemeny 1979). No detectable amounts of long-lived radioactive cesium or strontium escaped to the environment, although considerable quantities of each escaped from the fuel to the water of the primary system, the containment building, and the auxiliary building tanks.

The maximum estimated radiation dose received by any individual in the off-site population was 70 millirems. The collective dose to the surrounding 50-mile population was 2000 person-rems (about 0.8% of annual background radiation dose). These doses were mainly due to the biologically inert noble gases.

Extensive off-site monitoring, particularly for iodine-131 in milk, was conducted following the accident, but no significant levels of radioactive contamination were found in the environment around the facility. Fresh milk samples ranged from 16 to 41 pCi of iodine-131

per liter (Halsey 1980). This is well below the 12,000 pCi/L at which milk is considered unsafe to drink. Because of its 8-day half-life, background levels would be expected to be zero.

Thus, while this worst of U.S. power reactor accidents resulted in extensive contamination of facility buildings and equipment, off-site properties and agricultural lands were contaminated only by very small amounts of radioiodine which rapidly decayed. The noble gases, which escaped in large quantities, exposed the surrounding population to small amounts of radiation (external exposure and inhalation), but created no land contamination problems.

It is reassuring that off-site consequences of this accident were very much less, in terms of dose and contamination, than had been predicted for an accident of this kind. This discrepancy reflects the many conservative assumptions incorporated into reactor accident calculations.

### 2.3 NUCLEAR WEAPONS ACCIDENT ("BROKEN ARROWS")

The storage, distribution, and deployment of nuclear weapons since World War II has resulted in a potential for accidents, primarily related to transportation. Such accidents have been termed "broken arrows" by the Pentagon. The types of broken arrows include accidental release of unarmed nuclear weapons from aircraft, crashes of aircraft carrying nuclear weapons, missile propellant explosions involving nuclear warheads, and other transportation accidents or fires involving nuclear weapons. These accidents do not involve a nuclear explosion of the weapon, but only at most detonation or burning of conventional high explosives and dispersal of the fissionable material.

### 2.3.1 Extent of Contamination

The results of broken arrows include property losses, millions of dollars in cleanup operations, and the loss of kilogram quantities of uranium and plutonium. By their very nature (explosions, crashes), broken arrows have been associated with loss of life, but such deaths have not been a result of radiation exposure or from a nuclear explosion. To date no nuclear explosions have occurred from broken arrows. Part of the reason for the lack of a nuclear explosion lies in the design of the weapon. Conventional high-explosive detonators are arranged into an imploding configuration (lens) which focuses the energy of the blast to compress a mass of plutonium. The plutonium undergoes a fission reaction to produce the heat necessary to trigger the thermonuclear part of the weapon. The fission reaction will occur only if all segments of the lens detonate simultaneously. This is an unlikely consequence of a crash. Also, the weapon is protected from accidental triggering by an array of arming switches. Another safeguard against accidental arming is provided by the placement of switches, which must be activated simultaneously, in separate compartments. Any delay or out-of-order action cancels the arming sequence.

The area of contamination associated with broken arrows has varied from square meters to hectares. The most extensive area contaminated occurred near Palomares, Spain, where a mid-air airplane collision resulted in the contamination of several hundred hectares (Iranzo 1968). After the accident in Spain, 990 metric tons of contaminated soil (5000 barrels), from a contamination zone of 226 ha (Iranzo 1968), were removed and stored in South Carolina. According to Smith and

Lambert (1978), cleanup costs for this broken arrow exceeded \$205,000/ha (\$83,000 per acre). Two years later following an airplane crash in Thule, Greenland, 6712 m<sup>3</sup> of radioactive ice, snow, and crash debris collected from an approximately 22-ha (54 acre) contamination area (Hanson 1972) were similarly removed and shipped to South Carolina for long-term storage.

# 2.3.2 Source Terms and Hypothetical Case

The number of weapons accidents involving nuclear materials is not known with certainty. In two of the most comprehensive accounts one author (Hanauer, 1981) tabulates 28 broken arrows in the United States between 1950 and 1980, whereas the second author (Talbot 1981; Talbot and Dann 1981) lists up to 32 incidents (many of the disclosures concerning nuclear accidents have been in response to requests under the Freedom of Information Act). Hanauer (1981) speculates that, since the beginning of the atomic age, the number of accidents is probably nearer to 200. The weapons systems involved in most of the reported broken arrows have been aircraft carrying unarmed nuclear devices during training missions. There are reports of accidents involving missiles also. Because of the potential for aircraft accidents, in 1968 the Air Force stopped flying aircraft loaded with nuclear weapons on practice alerts and training missions.

## 2.3.3 Experience

The two most significant broken arrows occurred in 1966 and 1968.

The 1966 incident involved an American B-52 bomber and a KC-135 refueling aircraft that collided in midair at 3045 m elevation over

Palomares, Spain. Four unarmed nuclear weapons separated from the aircraft and fell to the ground. As a consequence, the conventional explosive component of two weapons detonated, resulting in damage to the weapons and a subsequent release of an aerosol of plutonium and uranium which contaminated the impact area (Iranzo 1968). A similar incident occurred two years later (Aarkrog 1971, 1977) when a B-52 aircraft trying to make an emergency landing at Thule Air Force Base in Greenland crashed on the ice in Bylot Sound. Again, the conventional explosive component of the nuclear weapon exploded causing the release of approximately 3 kg of plutonium over a 22-ha area. The latest broken arrow (1980) resulted from ignition of fuel vapors in a Titan II missile silo in Arkansas. A subsequent explosion threw the nuclear warhead several hundred meters from the site, but the warhead remained intact, and no radioactive material was released.

## 2.4 OTHER RADIOLOGICAL INCIDENTS

There is strong evidence that a large area (25-1000 km²) in the eastern Ural Mountains in Soviet Russia (in the Cheliabinsk Province between the cities of Kasli and Kyshtym near the Techa River) was heavily contaminated with fission-product radionuclides in the winter of 1957-1958 (the so-called "Kyshtym disaster"). The first public report of the Kyshtym incident was related by the dissident Soviet scientist Z. A. Medvedev (1976) and was later independently confirmed by an emigrant Soviet scientist living in Israel (Tumerman 1976). Medvedev (1976, 1979) maintained that the disaster involved a criticality which resulted from improper storage of medium— and

high-level radioactive waste from a plutonium production reactor.

Later reviews of available evidence led to speculation that the explosion was associated with radiochemical separations process wastes [(e.g., chemical explosion involving ammonium nitrate wastes (Trabalka et al. 1979; 1980)]. Whatever the cause, the accident was of unparalleled scale, and much can be learned from the Soviet response to it.

Official Soviet policy is to deny that a nuclear disaster ever occurred in the Ural Mountains, but independent and exhaustive reviews of the Russian radioecology literature (Medvedev 1979; Trabalka et al. 1980) have left little doubt that at least two lakes and an extensive area of land became highly contaminated, primarily with strontium-90, but also with cerium-144, cesium-137, ruthenium-106, and zirconium-95. In addition to this evidence, some confirmation has been found in declassified Central Intelligence Agency (CIA) documents released under the Freedom of Information Act (Medvedev 1979). There are very few first-hand eyewitness accounts recorded, and many descriptions of the situation and cleanup procedures are based on hearsay evidence. Nevertheless, a reconstruction of the postcontamination Soviet response can be made.

The explosion apparently resulted in some immediate deaths; the exact death toll is unknown. Radiation sickness apparently was observed in the general public downwind of the release, and there was an evacuation of residents from the most contaminated areas. Residential areas and personal belongings were burned to prevent citizens from either returning to their homes or removing contaminated

property. The highway between Cheliabinsk and Sverdlovsk, which traversed the contaminated area, was closed and then later reopened with signs advising travelers not to stop and to proceed at a high rate of speed. Lakes were closed to swimming and fishing, stocking of contaminated lakes with fish was suspended, hunting and trapping were prohibited, and stores which sold locally produced milk, meat, and foodstuffs were closed. Radiation monitoring of food and personal belongings was implemented. One eyewitness reported seeing fenced enclosures containing piles of topsoil. Medvedev (1979) speculates that these mounds resulted from the bulldozing or scraping off of topsoil. Rather than transfer or store the contaminated soil elsewhere, it was fenced off.

Trabalka et al. (1980) noted that approximately 30 small villages near the contaminated area disappeared from Soviet maps after 1958.

Some evidence from the declassified CIA documents indicates that larger cities were "decontaminated and covered with new layers of asphalt" (Medvedev 1979). Additional landscape changes since 1958 noted by Trabalka et al. (1980) were a network of canals, dams, and two new reservoirs constructed to alter drainage routes into the Techa River. The canals were used to divert water from the Techa River around the contaminated lakes, and the dams were constructed to prevent contaminated sediment and water from entering the Techa River from these lakes.

As stated before, much of this information is speculation based on evidence either gleaned from published Russian literature or on hearsay. Until the Soviets make an accounting, many details of the accident and countermeasures will remain unknown.

### 2.5 CHEMICAL LAND-CONTAMINATION INCIDENTS

After World War II the United States experienced a dramatic growth in the chemical industry which provided a boost in the nation's standard of living, but at the same time, provided a pervasive and insidious threat to the nation's health and safety through exposures to chemical toxins in both the workplace and in the home. By the 1970s, as many as 70,000 new chemical substances of commercial significance were in the marketplace, and sales in the industry exceeded \$112 billion in 1977 (Heritage 1978). However, only a few of the thousands of chemicals now produced have been adequately tested for possible chronic health and environmental effects.

It is beyond the scope of this document to adequately address the complex problem of land contamination via hazardous chemical wastes. A second report to follow (ORNL-6239) will examine hazardous chemical land contamination of land by nonradioactive chemicals in far greater detail. However, several example incidents will be presented here to point out the scope of the problem.

Land contamination via hazardous chemicals can range from several square meters from a local spill to contamination of thousands of square kilometers of land from a major incident. Hazardous chemicals have contaminated land, rivers, lakes, groundwaters, and, indeed, the entire global ecosystem as in the case of polychlorinated biphenyls (Risebrough et al. 1968). Railway and tanker truck accidents involving hazardous chemicals have prompted evacuation of neighborhoods and entire towns. Major spills or seepage from illegal or improperly

constructed land disposal sites have also resulted in the evacuation or relocation of entire towns. Some of the most extensive land contamination events include the following:

- In July 1976 a chemical reactor used in the manufacture of 2,4,5-trichlorophenol malfunctioned and vented its contents, including from 2 to 3 kg of 2,3,7,8-TCDD (dioxin) near Seveso, Italy (Hay 1976a,b,c; Bisanti et al. 1981; Kriebel 1981). Over 111 ha (275 acres) were contaminated, but it was not confirmed that TCDD had been released until two weeks after the accident. Over 30 people showed symptoms of chloracne (disfiguring burns and skin rash), and over 1000 deaths of birds and domestic animals were reported. Eventually, over 800 people were evacuated from the most contaminated zone (many permanently). TCDD is a known teratogen, and 29 Italian women who were exposed in the accident eventually obtained abortions. Decontamination of the most extensively contaminated area included clearing and burning of residences, structures, and vegetation and removal of topsoil at a reported cost of \$45 million (Renzoni 1977).
- In March 1966 production of the pesticide Kepone was consolidated at Allied Chemical's Semi-Works plant at Hopewell. Virginia. on the banks of the James River. Operation of the Kepone plant continued until 1974 when Kepone production was taken over by Life Science Products Company (LSPC) under agreement with Allied When the LSPC plant closed in July 1975, approximately Chemical. 18,000 kg (40,000 lbs) of Kepone had contaminated roughly 104 river kilometers (65 miles) of the James River or approximately 5000 ha [60 million square yards of river bottom (Steele and Gilley 1981)]. The result of such a widespread contamination was closing of the James River from Richmond, Virginia, to the mouth of the Chesapeake Bay to sport and commercial fishing. Estimated cost for proposed cleanup of the James River exceeds \$3 billion (EPA 1978). In addition to the river contamination, there was extensive soil contamination near the plant. Dust collected approximately one block from the LSPC plant ranged from 1-40% Kepone by weight; soils near the plant were 1-2% Kepone; and at 1 km from the plant, Kepone levels in soil were 2-6 ppm (Huggett and Bender 1980).
- In the 1920s a three-block tract of land, Love Canal, on the eastern edge of Niagra Falls, New York, was turned into a municipal and industrial chemical dumpsite. The Hooker Chemical Company, which owned and operated the Love Canal dumpsite, covered it with earth and sold it to the city of Niagra Falls for one dollar in 1953. In the years immediately following that time, approximately 100 homes and a school were built at the site. In the summer of 1982 after a record amount of rainfall, roughly 82

different compounds, 11 of them suspected carcinogens, began to percolate up through the soil into the backyards and basements of the Love Canal community. Trees and gardens began to die, and "everywhere the air had a faint, choking smell" (Beck 1979). Eventually, the entire community (221 families) was evacuated from the Love Canal site. The cost to the state of New York of purchasing the evacuated homes was \$7 million alone. The residents are now being closely watched for increased incidence of birth defects, miscarriages, and cancer.

In 1971 a waste hauler soaked a horse arena in Moscow Hills, (near Times Beach) Missouri, with 7500 L (2000 gal) of waste oil to control dust for an upcoming horse show (Sun 1983a). Almost immediately, hundreds of birds, dozens of dogs and cats, numerous flies and rodents, and more than 60 horses died. A young girl, Andrea Piatt, also became sick and was permanently disabled (Heritage 1978). Eventually, it was learned that the waste oil was heavily contaminated with TCDD (dioxin) from a waste tank at a chemical plant which produced hexoachlorophene. The horse arena spraying was, however, only the tip of the iceburg. Approximately 70,000 L (18,500 gal) of the contaminated oil was sprayed around the state of Missouri for dust control by the same waste hauler. Officials have confirmed 14 contaminated sites, and suspect roughly 100 more. At Times Beach, dioxin contamination of up to 100 ppb in soil is widespread. Roughly 800 families are pondering their future finances and health, and state and federal officials lack clear, precise answers to the problems of cleanup (Sun 1983b).

The above examples are but a few of the most publicized incidents. Unfortunately, hundreds more incidents have occurred, and thousands more are likely to occur in the future. Both individuals and whole communities can be affected, and the eventual cost in terms of agricultural and property losses, medical care, and land cleanup and reclamation are exceedingly high.

### 3. IMPACTS

### 3.1 EXTERNAL RADIATION DOSES

At one year following serious reactor accidents or nuclear weapons detonations, only about a dozen radionuclides remain on contaminated land to produce significant external doses to exposed individuals. Two pathways of external exposure are associated with contaminated ground. There is direct exposure from gamma rays from radionuclides on or in the ground, and there is exposure from immersion in contaminated air into which radionuclides have been resuspended from soil surfaces. The latter pathway leads to relatively small external exposures because only  $10^{-5}$  to  $10^{-9}$  of the soil surface activity becomes resuspended in air, depending on time elapsed since the initial deposition (U.S. NRC 1975).

Table 3.1 gives dose rates to the total body from radionuclides on the soil surface and in the air above the soil. These dose rates are conservative in that no structural shielding is assumed, a smooth ground surface is assumed, and no provision is made for weathering effects. Over time, radioactivity on surface soil is leached into the soil, thus lowering exposure rates due to the shielding properties of the soil. Of course, radioactive decay also will reduce dose rates, but with the exception of cobalt-60, ruthenium-106, cesium-134, and cerium-144, the radionuclides in Table 3.1 are relatively long-lived.

The effect of weathering of radionuclides deposited on soil surfaces has been demonstrated in the case of fallout, and all radionuclides penetrate into the soil with time. The actual rate of

Table 3.1. External dose rates to the total bodya

	Instantaneous dose rate		
Radionuclide	Contaminated ground <u>b</u>	Contaminated air (mrems/y/mCi/cm²) (mrems/y/mCi/cm³)	
Co-60	2.5 x 10 <sup>6</sup>	1.5 x 10 <sup>10</sup>	
Ru-106, Rh-106	2.3 x 10 <sup>5</sup>	1.1 x 10 <sup>9</sup>	
I-129	$2.6 \times 10^4$	$4.9 \times 10^{7}$	
Cs-134	1.7 x 10 <u>6</u>	8.5 x 10 <sup>9</sup>	
Cs-137,Ba-137m	6.5 x 10 <sup>5</sup>	$3.2 \times 10^9$	
Ce-144	2.5 x 10 <sup>4</sup>	1.0 x 10 <sup>8</sup>	
Pu-238	$8.0 \times 10^{2}$	$4.3 \times 10^{5}$	
Pu-239	3.6 x 10 <sup>2</sup>	4.3 x 10 <sup>5</sup>	
Am-241	$3.2 \times 10^4$	1.0 x 10 <sup>8</sup>	
Cm-242	9.1 x 10 <sup>2</sup>	$4.9 \times 10^{5}$	
Cm-244	8.1 x 10 <sup>2</sup>	3.8 x 10 <sup>5</sup>	

aSource is Kocher (1979).

 $\underline{b}_{\mbox{\scriptsize Radioactivity}}$  is on soil surface and no shielding or surface roughness effects are assumed.

movement depends on the chemical form of the radionuclide, chemical and physical properties of the soil, and external environmental factors, such as precipitation and temperature fluctuations. In general, weathering will reduce external exposures from soil surface contamination to about one-half initial exposure rates in two years. Subsequent weathering, however, will not reduce exposures appreciably because deposited radionuclides will tend to reside in the top few centimeters of soil (U.S. NRC 1975), and radiological decay will become the dominant means of exposure reduction.

For both serious reactor accidents and nuclear weapons fallout, cesium radionuclides, ruthenium-106, cerium-144, and cobalt-60 will provide most of the external exposures at one year after initial deposition. After about five years, external exposures will be due mainly to cesium-137. The transuranic radionuclides (plutonium, americium, curium) and iodine-129 contribute relatively little to external exposures because they are produced in relatively small quantities. These alpha and beta particle emitters are more important in terms of internal doses (inhalation and ingestion) due to their long half-lives and energy characteristics.

Radionuclides such as tritium (H-3) and carbon-14 that are produced by nuclear explosions, but not by serious light-water reactor accidents, do not contribute very much to external exposures because they are beta emitters. Like iodine-129 and the transuranic radionuclides, they are more important in internal-dose pathways.

The external dose from fallout deposited after surface detonation of a one-megaton weapon, and the area involved, may be estimated from

Table 2.2. At one year following detonation, about 94% of the fallout radioactivity will have decayed, leaving residual doses given in Table 2.2. An estimate of external doses at one year is given in Table 3.2. About 5% of the affected area has exposures over 500 millirems/year, which is the current upper limit of nonoccupational annual exposure for individuals in the nuclear power industry. Internal exposures from inhalation of resuspended particulate matter and ingestion via food chains would increase total doses. About 70% of this external exposure at one year following deposition is due to ruthenium-106 and cerium-144. Because both of these radionuclides have half-lives on the order of one year, they will decay such that after five years the total external exposure rate will be only 4% of the rate at one year. At this time, cesium-137 will account for about 80% of the external dose and will continue to provide most of the external dose until it decays.

In the case of multiple weapons detonations, or a nuclear war, relatively high external exposures would prevail in the northern hemisphere. Given the average deposition of fallout radionuclides given in Table 2.4 for a 10,000 megaton nuclear war, the external dose outside of the combat areas at one year would be 770 millirems/year, largely due to cesium-137. At five and ten years following deposition, the external dose would be 211 and 159 millirems/year, respectively, due to radiological decay of ruthenium-106 and cerium-144.

An estimate of external doses and the areas involved at one year following the most serious reactor accident are given in the WASH-1400 report (U.S. NRC 1975), and are reproduced in Table 3.3. At one year,

Table 3.2. External doses from fallout at one year following detonation of a one-megaton weapon

Area (km <sup>2</sup> )	Dose (mrems/y)	
900	>1,000	
5,500	500 to 1,000	
5,500	250 to 500	
29,100	60 to 120	
88.700	12 to 30	

Table 3.3. Estimated external dose rates at one year following deposition of radioactivity from the most serious reactor accidenta

Area (km²)	Distance downwind (m) b	Dose (mrems/y)	
650	0 to 50,000	>1000	
425	50,000 to 67,500	500 to 1,000	
1,400	67,500 to 100,000	250 to 500	

 $\underline{a}A$  core meltdown with a steam explosion and loss of containment, probability is 9 x  $10^{-7}$  per reactor year (USNRC 1975).

 $^b\mbox{Assume}$  all radioactivity blows downwind in a 22.5° sector segment.

the external dose is due primarily to cesium-134 (60%), cesium-137 (21%), and ruthenium-106 (16%). After five years, the dose rate is down by about a factor of 3 due to decay of ruthenium-106 and cesium-134. By ten years, the dose rates will be about a factor of 5 less. At this time an area of about 300 km<sup>2</sup> would have external exposures exceeding 500 millirems/year.

If weathering of particulates deposited on the soil surface is considered, all of the above external exposures at one year might be reduced by about a factor of 2 in two years in areas receiving moderate rainfall. Further reductions would result from ground roughness and structural shielding.

## 3.2 HUMAN HEALTH EFFECTS

Results from biological experiments, a few accidents, and experience with nuclear weapons dropped on Japanese cities in World War II have served to document acute effects of high doses of ionizing radiation. Biological effects ranging from radiation sickness to death are associated with whole-body doses exceeding 100 rems given over a short period of time.

Estimates of human health effects from exposure to low levels of radiation can be made for any level of exposure. The health effects expected from exposures to low levels of radiation, such as those associated with contaminated land considered in this study, may include small increases in the number of cancers among exposed populations and potential genetic effects that may be expressed in future generations.

Estimates of risk factors for exposure to radiation, both for individuals and populations, are available in a number of publications. The National Academy of Sciences issued a report from the Advisory Committee on Biological Effects of Atomic Radiation called the <u>BEAR Report</u> (NAS 1960). In 1972, the National Academy of Sciences Advisory Committee on the Biological Effects of Ionizing Radiation issued a report commonly referred to as <u>BEIR I</u> (NAS 1972).

The committee later was asked to review the risk estimators for health effects from radiation, and the results of that review were issued in the 1980 report referred to as <u>BEIR III</u> (NAS 1980). Other groups of experts also have published risk estimators for radiation exposure; for example, the International Commission on Radiological Protection (ICRP) and the National Council on Radiation Protection and Measurements (NCRP) have both studied radiation effects. The ICRP issued <u>Publication 26</u> in 1977 (ICRP 1977). In the same year a United Nations study group (U.S. Scientific Committee on the Effects of Atomic Radiation, or UNSCEAR) published an extensive report that included estimates of risks of cancer from ionizing radiation (UNSCEAR 1977).

### 3.2.1 Cancer

The cancer risk estimators developed by various organizations are presented in Table 3.4. The values presented represent the estimated range of added cancer deaths (above the normal deaths from such cancers) in a population of one million people exposed to 0.5 rem/y external gamma radiation. These estimates were calculated from data provided by the Nuclear Regulatory Commission (American Cancer

Table 3.4. Estimates of cancer mortality in a population of one million exposed to 0.5 rem of radiation.

Source	Additional (	fatal cancers <u>b</u>	
BEIR (1980)	35-	35-85	
ICRP (1977)	50		
UNSCEAR (1977)	40-	-90	

<u>a</u>All three groups estimated cancer mortality from radiation-induced cancers. The American Cancer Society has stated that only about one-half of all cancer cases are fatal. Thus, to estimate total incidence of cancer, the above values should be multiplied by 2.

b"Additional" means above the normal cancer rate.

Society 1978) to indicate the range of health effects predicted for given radiation exposures.

Comparison of the three sets of values in Table 3.4 indicates that there is general agreement among health-effects estimates developed by the three studies. In application of the risk estimators, care must be taken to identify the limitations of each study. The UNSCEAR and ICRP approaches provide age-weighted, average-lifetime risk factors. The UNSCEAR report uses absolute risk, or the number of expected cancer cases that will result from exposure of a given population. The <a href="BEIR III">BEIR III</a> report uses relative risk, which is the ratio of incidence in an exposed population to the incidence in a control population.

The American Cancer Society (1978) indicates that about half of all cancer cases are fatal. Thus, the numbers in Table 3.4 can be multiplied by 2 to estimate total incidence (not just fatalities, but also injury and illness). These cancers would be in addition to those normally expected in a population. According to the American Cancer Society, the individual risk of getting cancer for any member in the population is 1 chance in 4. The cure rate for these cancers ranges from 90 to 5%, depending on the type of cancer.

The values given Table 3.4 are the best estimates that can be provided by national and international experts in radiation biology and protection. Press releases have publicized radiation-effects studies in which the authors have indicated that the risk is much higher than represented by the estimates given in Table 3.4. It should be noted, however, that the National Academy of Sciences (BEIR Advisory Committee) did take these other studies into account in setting of risk estimators.

There have been no direct measurements of increased cancer for low-level radiation exposures (1-5 rems). Data exist only for much higher exposures (typically 100 rems and above). Risks at lower doses have been estimated by assuming that the same dose/health-effects relationship applies to low doses as to high doses, and then linearly extrapolating from data taken at higher dose levels to these low-dose levels.

Organ-risk estimators have been developed in UNSCEAR and the National Academy of Sciences reports for radiation exposure of organs where radionuclides selectively deposit. Risk factors given in <a href="https://doi.org/10.25">BEIR III</a> for selected organ doses are listed in Table 3.5. An estimate of the probability of injury to the individual as a result of a particular dose can be obtained by multiplying the estimated dose by the appropriate risk factor.

In the <u>BEIR III</u> report, two different risk-projection models were used. The absolute-risk projection model was used to estimate the incremental risk due to radiation exposure, as the arithmetic difference between the risk to individuals in the exposed group and those in a control group receiving no radiation exposure. The relative-risk projection model was used to express risk as the ratio of the risk of the exposed group to that of the unexposed group. The risk factors in Table 3.5 are based on the absolute-risk projection model for the specific organ doses. The risk factor for whole-body exposure is based on both projection models averaged over both age and sex.

Another manner of looking at risk from radiation is that the induction

Table 3.5. Cancer mortality for selected organ doses₫

· .	Lifetime risk of mortality		
Organ exposed/ type of cancer	Per mrad	Per mrem	Type of radiation <u>b</u>
Whole body/all cancers	120 x 10 <sup>-9</sup>	120 x 10 <sup>-9</sup>	Low LET
Lung/lung cancer	900 x 10 <sup>-9</sup>	99 x 10 <sup>-9</sup>	High LET <u>C</u>
Liver/liver cancer	300 x 10 <sup>-9</sup>	30 x 10 <sup>-9</sup>	High LET <u>C</u>
Bone surface/bone cancer	140 x 10 <sup>-9</sup>	1.4 x 10 <sup>-9</sup>	High LET <u>C</u>
Bone volume/bone cancer	20 x 10 <sup>-9</sup>	$2.0 \times 10^{-9}$	High LET <u>⊆</u>

aSource is BEIR III report (NAS 1980).

 $\underline{b}$ LET = linear energy transfer (kiloelectron volt per micrometer in water). Low LET radiation includes x-rays, gamma-rays, and beta particles. High LET radiation includes alpha particles and neutrons.

<u>CA</u> quality factor of 10 for alpha particles (high LET radiation) has been used. However, if the ICRP-30 dose calculational methods are used, a quality factor of 20 for alpha particles should be applied. The absorbed doses are multiplied by the LET-dependent quality factor to obtain (for radiation protection purposes) a quantity that expresses the effectiveness of the absorbed dose on a common scale for all ionizing radiations.

of cancer results in a life shortening that would not take place otherwise. When evaluated by the same method, average background radiation (about 0.1 rem/y) results in an estimated life shortening of eight days (Cohen and Lee 1979).

### 3.2.2 Genetic Effects

Genetic effects are estimated to all subsequent generations as a result of an annual exposure at a particular radiation level. The risk factor used to estimate the total of all genetic effects was taken from BEIR III to be from 60 to 1100 total cases of genetic disorder per 1,000,000 live-borne offspring for each rem of dose from low LET radiation (x-rays, gamma-rays, and beta particles), and from 180 and 3300 total cases of genetic disorder per 1,000,000 live-borne offspring for each rem of high LET radiation (alpha particles and neutrons). The dose involved here is the gamete dose, which is calculated from the gonadal dose of the exposed individuals, their age and sex distribution, probabilities of having children, and from adjustment factors of 0.82 and 0.18 for male and female exposures, respectively, to account for the spermatogonia and oocyte mutational sensitivities.

The risk factors given above do not take into account age grouping or sex, but instead, represent average population risk estimators. For planning purposes, average estimated risks appear to be acceptable; however, for site-specific risk analyses, age- and sex-specific risk factors should be used in conjuction with demographic data for the site.

It should be noted here that no increases in genetic effects (mutations) have been observed in offspring of survivors of the

Hiroshima and Nagasaki weapon blasts (NAS 1980; Grosch and Hopwood 1979). Therefore, the genetic effects estimates are based only on theory, and not experience.

#### 3.3 ENVIRONMENTAL PATHWAYS TO MAN

In emergency situations, the food-chain transport of toxic substances to humans is of secondary importance to other more direct modes of exposure such as inhalation, drinking water, or direct contact with the skin. This is because the food-chain transport of contaminants along the pathways: soil-to-plant-to-human, or soil-to-plant-to-animal product-to-human, involves one or more transfers across biological membranes where the potential for discrimination against contaminant transfer exists. Environmental conditions, such as soil type and climate, can modify the potential of toxic materials for food-chain transport. Hazards and environmental impact on wildlife, domestic animals, and humans can arise in two ways:

- shortcuts in the food-chain transfer of a toxic substance can bypass opportunities for discrimination in biological transfer and
- the chemical properties of a compound, coupled with its biochemical pathways, can result in its bioaccumulation or bioconcentration in plants or animals.

### 3.3.1 Transfer to Plants

The entry of a food-chain contaminant into plants and water is generally the first step in its transfer to livestock and humans. The pathways of plant contamination considered here are root uptake from soil and direct deposition on plant surfaces.

3.3.1.1 Root Uptake. The conventional method for predicting the transfer of contaminants from soil to plants involves the calculation of a "concentration ratio" (CR value) where:

CR = concentration per gram in plants/concentration per gram in soil.

Historically, the CR value has been used in models designed to predict the food-chain transport and subsequent radiological impact of radioactive pollutants (e.g. see Ng 1982). A recent review and analysis of concentration ratios, determined from both laboratory and field experiments, for most long-lived radionuclides can be found in Baes et al. (1984). There are numerous soil factors (such as texture, pH, or organic matter content) which are known to affect the root uptake of various radionuclides.

Similarly, soil properties can affect the extent of plant uptake of toxic organic wastes from soil, although there is generally little data on concentration ratios for organic chemicals. This is partly due to the large number of organics to be investigated; there are about 63,000 chemicals in common use, many of them organics (Maugh 1978). By comparison, there are less than four dozen radionuclides and potentially toxic heavy metals that are of potential concern from the standpoint of human exposure through food chains.

Several generalizations can be made concerning the entry of inorganic and organic contaminants into plants from the soil.

• Environmental conditions or biological processes that increase the water solubility of a radionuclide or an organic chemical will tend to increase its entry into vegetation via plant roots.

The water solubility of a contaminant is of paramount importance in determining the extent of uptake from soil by plant roots. It has long been recognized that factors which affect the water solubility of both inorganic and organic compounds also affect uptake by plants. Conversion of inorganic chemicals to insoluble forms by amendments to contaminated soils can reduce plant uptake. For example, plant uptake of strontium-90 from some soils can be reduced by large applications of phosphorus resulting in the formation of insoluble strontium phosphates (Francis 1978).

The affinity of contaminants for soil and their equilibrium distribution between the soil and the soil water is expressed in most food-chain models as a distribution coefficient or  $K^{\hat{\mathbf{d}}}$  value, where

K<sup>d</sup> = concentration per gram in soil/concentration per milliliter solution.

The K<sup>d</sup> value is correlated with the soil-to-plant concentration ratio for many inorganic elements (Baes et al. 1984). For example, the sorption of transuranic elements to soil follows the ranking plutonium > curium >> neptunium and is inversely related to plant uptake (Watters et al. 1980).

Polarity (as an antecedent to water solubility) is of considerable importance in determining the plant uptake of organic contaminants. Plant roots do not tend to exclude small organic molecules (molecular weight less than 500) unless they are nonpolar (Nash 1974). Nonpolar organic contaminants tend to adsorb on the external root surface rather than enter the plant.

Synthetic and natural organic chemicals have the potential to alter the water solubility of inorganic elements, and thereby, increase their uptake by plant roots. Careful attention must be paid to soil amendments which are used in reclaiming contaminated land. For example, organic complexation of heavy metals and some radionuclides can result in order of magnitude increases in water solubility and increased plant uptake. Chelating agents, like diethylene-triaminepentaacetic acid (DPTA), which may be present in industrial wastes, can greatly increase the uptake of transuranic elements by plants (Ballou et al. 1978).

 The uptake of radionuclides and organic chemicals is greatest in sandy soils having low clay content and low organic matter.

Many inorganic elements and many organic compounds tend to sorb onto soil clays and soil organic matter. This subsequently reduces their availability and concentration in the soil solution. Sorption onto clay may arise from the internal incorporation into the mineral lattice (as is the case for cesium-137 binding with illite clay) or simple adsorption due to differences in surface charge. In either case, the presence of clay in soil will help to pull many different kinds of contaminants out of the soil solution, and thereby, reduce their uptake by plants. Organic matter has a similar effect. The presence of organic matter in soil is especially important to the binding of nonpolar organic contaminants.

For organic compounds only, volatilization of the chemical from soil can be a far more important plant contamination pathway than uptake from soil via plant roots.

Under field conditions, it is usually very difficult (and sometimes impossible) to quantify the relative contribution of various pathways (root uptake, splashing, resuspension, volatilization, etc.) through which an organic chemical reaches plant foliage. It has been shown in laboratory studies that the volatilization of DDT, dieldrin, endrin, and heptachlor (all insecticides) contributes more to the contamination of plants than uptake via plant roots (Nash 1974).

• The amount of uptake and its apportionment among roots, stems, leaves, and edible parts will vary with the contaminant studied, its metabolism in the plant, plant age, and plant species.

The distribution of contaminants in plants can be uniform, acropetal (increasing concentration gradient from roots to foliage), or basipetal (increasing concentration gradient from foliage to roots). The type of distribution depends upon plant metabolism. The long-lived radionuclide technetium-99 has an acropetal distribution in most plants, while plutonium-239 has a basipetal distribution. The distribution of a pollutant in crops will govern crop use when grown on contaminated lands. If the distribution of a pollutant in a crop is basipetal, then some aboveground parts may be considered acceptable for consumption even when the root or tuber is not acceptable.

 In the absence of significant root uptake and translocation, it is still possible to have accumulation of contaminants in root crops because of surface adsorption.

Various heavy metals, radionuclides, and organic pollutants are known to concentrate on the surfaces of plant roots. For example, CR values for mercury uptake by crops are approximately a factor of 6

greater for roots than for aboveground foliage (John 1972). In carrots grown on soil treated with the organic insecticides aldrin and heptachlor, most of the insecticidal residues are associated with the carrot epidermis or peel (Lichtenstein et al. 1965). Nonpolar organic molecules do not easily pass through the root epidermis; instead, they adsorb on the root surface (Nash 1974).

 The persistence of an organic chemical in soil will, to a large extent, govern concern about its potential for uptake by plant roots and subsequent transfer through the food chain.

Organic contaminants that are quickly destroyed (half-time in soil less than four days) by biodegradation, photolysis, or other means are of little concern to food chains because more than 90% of the contaminant will be degraded within a fraction of a plant's growing season (Gillett 1983). On the other hand, organics with degradation half-times longer than 14 days should be considered as possible candidates for uptake via plant roots because more than 10% of the initial soil concentration may persist for a substantial fraction of a growing season (Gillett 1983).

3.3.1.2 Direct Deposition. Direct deposition of contaminants onto food or forage crops can be an important shortcut in the food-chain transfer of contaminants because the opportunity for discrimination at the point of the plant root is bypassed. Direct absorption of many pollutants through the leaves of plants is prevented by the lipid properties of the plant's epidermis. Generally speaking, the leaf has evolved for the function of gas exchange and photosynthesis, and not nutrient absorption. Nonetheless, leaves can adsorb or absorb some

inorganic elements and organic compounds when they are applied to foliar surfaces. Oils and emulsions can increase the penetration of organics through the leaf surface (Prendeville and Warren 1975), and certain properties of leaves (i.e., waxy cuticles, leaf hairs, shape, and texture) can make them efficient traps for small particles and their associated contaminants. Experiments in which plants were exposed to transuranic aerosols have shown that foliar accumulation and translocation to seeds can be just as important as uptake via plant roots (Cataldo et al. 1980).

It would at first seem that it makes little difference to food-chain transfer whether a contaminant is incorporated into the plant or merely present on the plant's surface. However, contaminants that are biologically incorporated in plant tissue have the potential for forming associations with products of metabolism which may make them more or less available for gastrointestinal absorption by animals and man. Furthermore, the direction of change in availability depends upon the pollutant considered. For example, the incorporation of plutonium into plant tissues slightly increases its availability for food-chain transfer, while incorporation of technetium into plant tissue produces an opposite result (Sullivan et al. 1979).

Plant uptake of contaminants depositing in aerosols is treated very simply in most mathematical models used to predict food-chain transport. Vegetation density, the fraction of the deposit intercepted, and the removal half-time from the vegetation are the key parameters that must be quantified to predict concentrations in an exposed crop. One complicating aspect of modeling deposition is that fractional

interception of a depositing aerosol depends on the vegetation density. Baes et al. (1984), in a recent review and analysis of food-chain model parameters, have presented theoretical curves relating fractional interception to biomass density for different types of vegetation.

Several generalizations can be made concerning the contamination of plants by direct deposition:

 From the standpoint of food-chain transfer, direct deposition of contaminants onto plants is important for those food crops for which foliage or exposed parts are consumed directly by people (for example, lettuce, cabbage, broccoli, and leafy vegetables).

Thick peels, shells, or pods will protect some food crops from exposure to contamination by direct deposition. Standard methods of food preparation, such as washing, peeling and trimming, and cooking, have the potential to reduce the levels of contaminants present on crops (Geisman 1975). Cooking, in particular, has the potential for degradation of organic contaminants when they are not thermally stable. Cooking, however, has the potential to release volatiles into the air and increase inhalation exposures.

 In most cases, deposits of large particles on vegetation will be rapidly lost through exposure to natural meteorological processes, but submicron particles may be retained for long periods of time, depending upon the properties of the leaf surface.

The weathering of deposits by precipitation and wind will cause a loss of contaminants from vegetation with time. The amount of time required to reduce the initial concentration on vegetation by 50% is

known as the weathering half-time. Large particles greater than the size of fine sand (0.1 mm or larger) are usually easily removed from vegetation by natural weathering. However, submicron particles are not easily dislodged from leaf surfaces, and contaminants associated with such particles can be retained for long times by the plant (Cataldo et al. 1981).

 There are at present no adequate simple models for predicting absorption of volatile organics by foliage.

#### 3.3.2 Transfer to Animals

Several important food-chain transfers to animals must be considered in evaluating the potential agricultural uses of contaminated lands. These transfers are particularly important in those cases where contamination is so extensive as to preclude cleanup, by topsoil removal or other means, of all areas except those that are the most heavily affected. The general food-chain pathways of interest are soil-to-forage-to-meat or milk or eggs, and soil-to-meat or milk or eggs. The relative importance of these transfers will depend upon environmental conditions (e.g., soil type), livestock grazing habits, and animal management practices.

Compilations of data on the transfer of various long-lived radionuclides from contaminated forage to beef, pork, lamb, chicken, eggs, and milk are available in the published literature (Ng 1982; Baes et al. 1984). Based on these compilations, the annual effective whole-body dose (weighted sum of doses to individual organs) from ingestion of foods grown on land contaminated to a level of 1  $\mu$ Ci/m<sup>2</sup>

strontium-90, cesium-137, and plutonium-239 can be calculated (Table 3.6). A level of 1  $\mu$ Ci/m<sup>2</sup> strontium-90 deposited on the ground would occur within roughly 100-300 km downwind of ground-zero for a nuclear weapon's detonation and would be equivalent to an external gamma dose rate of 100 roentgens (R)/h (1 R = 2.58 x 10<sup>-4</sup> C/kg in air) or greater (Bondietti 1982).

In our calculations radioactivity is initially applied to the land surface, but is plowed to a depth of 15 cm. Surface activity at a level of 1  $\mu$ Ci/m<sup>2</sup> will be reduced to roughly 5 pCi/g of soil after plowing to a depth of 15 cm. As can be seen from Table 3.6, vegetables and produce grown on contaminated land will account for most of the ingestion dose, followed by milk and eggs, and beef, pork, and chicken. That is, while contamination levels in vegetables and produce may be prohibitive, production of milk, eggs, and meats may be an alternative land use.

Organic chemicals must persist in the environment before they are candidates for food-chain transfer. Persistent lipophilic organic contaminants characteristically accumulate in animal fat, and data compilations are available for the transfer of some organic chemicals from contaminated diets to animal fat (Garten and Trabalka 1983; Kan 1978). However, no data on food-chain transfer exist for a very large number of organic contaminants which might be encountered in the environment, and predictions of the food-chain accumulation of organic chemicals in animal fat, based on their physicochemical properties, are characterized by large uncertainties (Garten and Trabalka 1983).

Table 3.6. Annual effective whole-body dose commitment from foodstuffs grown on land contaminated to a level of l  $\mu\text{Ci/m}^2$  on the soil surface

Nuclide	Dose <u>b</u> (mrem/y)	Percent contribution		
		Vegetables <u>C</u>	Milk and eggs Meats <u>d</u>	
Sr-90	20	80	18	2
Cs-137	1.0	60	21	19
Pu-239	0.009	100	<0.1	<0.1

aBased on Baes et al. (1984); Ng et al. (1982); and Shor et al. (1982), assuming all food, including cattle feeds, grown on site.

 $\underline{b}$ Effective whole-body dose via ingestion (Till and Meyer 1983). The effective whole-body dose is the weighted sum of doses to individual organs.

 $\underline{\underline{C}}$ Assuming the annual U.S. average consumption rate of leafy vegetables, "exposed" and "protected" produce, and grain given by Nelson and Yang (1984). Root uptake pathways only are considered.

 $\underline{\mathbf{d}}$ Beef, pork, and chicken.

Based on current knowledge of the food-chain transfer of inorganic pollutants (principally radionuclides) to animals, there are several important generalizations of which one should be aware in deciding on the use of contaminated agricultural lands:

Inorganic and organic substances differ in their metabolism, tissue distribution, and excretion from the body. This makes it impossible to accurately predict the potential food-chain bioaccumulation of complex mixtures of contaminants without specific information on the animal species and the particular contaminants involved.

Data on metabolism and tissue distribution from animal feeding studies are essential to making any confident assessment of the importance of food-chain transfer to animals. This is because various pollutants can differ markedly in their fate and distribution in animals. The metabolism of inorganic elements has been extensively studied, and their distribution within animals can be forecast with great confidence. For example, radiocesium is distributed throughout muscle; strontium and many transuranic elements are accumulated in bone; radioiodine accumulates in the thyroid; and heavy metals tend to accumulate in kidneys or the liver. Although many organics accumulate in animal lipids, some of the more polar compounds can also be found in liver and kidneys. Information on excretion from livestock tissues is of critical importance to predictions of food-chain transfer because the excretion rate will control the levels to which contaminants are bioaccumulated.

Ingestion of contaminated soil can be more important, or just as important, to the food-chain transfer of contaminants as ingestion of contaminated forages. Ingestion of soil by livestock and birds is an important way in which the food-chain transfer of contaminants can be shortcut. On the average, cattle ingest 0.5 kg of dry soil per animal per day, although this amount can vary considerably depending upon range conditions (Zach and Mayoh 1984). Ingestion of contaminated soil is most important for those elements which exhibit low soil-to-plant transfer (e.g., thorium, uranium, and the transuranic elements). Animals can assimilate inorganic elements, like lead (Dacre and Ter Haar 1977), and organic substances, like dioxin (McConnell et al. 1984), directly from contaminated soils. Because of the tendency of animals to accidentally or intentionally ingest soils, the maintenance of animals on stored feed may not prevent the transfer of contaminants to animals confined to contaminated lands.

 Organic substances of most concern for bioaccumulation in terrestrial animals are lipophilic compounds that are environmentally persistent.

Environmental half-times for many organic contaminants in soil are highly dependent upon soil type, climate, temperature, organic matter content, microbial activity, and other environmental conditions (Kearney et al. 1969). In general, organic compounds with environmental half-times in soil of less than four days and  $\underline{n}$ -octanol-water partition coefficients less than 300 are of no ecotoxicologic concern from the standpoint of food-chain transfer (Gillett 1983). The tendency of an organic compound to bioaccumulate from the diet into animal lipids is correlated with its distribution in a two-phase liquid system of  $\underline{n}$ -octanol and water (Kenaga 1980). On the other hand, organic

compounds with environmental half-times in soil greater than 14 days and  $\underline{n}$ -octanol-water partition coefficients ( $K_{OW}$ ) greater than 3000 (e.g., the polycyclic aromatic hydrocarbons) are of great concern with respect to potential food-chain transfer. Certain classes of organic compounds, like organochlorines, tend to be very stable and persistent in the environment, while other organics, like organophosphates, are degraded within a matter of weeks or months. Estimated environmental half-times in soil for a variety of organic chemicals have been summarized by Trabalka and Garten (1982).

 Agricultural practices which increase the mobility of contaminants in soil may also increase the potential exposure of livestock.

Remedial actions on contaminated lands may involve chemical treatments to accelerate the downward movement of contaminants out of the plant rooting zone, or chemical treatments designed to accelerate the uptake and removal of contaminants in plant biomass. For example, additions of chelating agents to soil will increase plant uptake of transuranic elements, but chelated transuranics absorbed upon ingestion by rats are excreted with little tissue retention (Ballou et al. 1978). Chemical amendments which increase plant uptake may not necessarily increase retention in animal tissues, but amendments that increase plant uptake will increase the level of exposure to the animal's gastrointestinal system.

# 3.4 ECOLOGICAL CONSEQUENCES

Most of the knowledge of the effects of ionizing radiation on plants of natural terrestrial ecosystems is based on point-source

exposures of a variety of ecosystems and on laboratory and field studies of the radiation sensitivities of species from these ecosystems.

Similarly, our knowledge of the effects on natural populations of animals is based on research on animals living in areas contaminated by nuclear wastes, or wild animals exposed to point sources. Relatively accurate predictions of plant radiosensitivity can be made from characteristics of the plant nucleus and radiation exposure or dose (Sparrow et al. 1963a). Details of radiation effects on plants, terrestrial ecosystems, and natural animal populations are available in reviews, notably Whicker and Fraley (1974) and Turner (1975). In contrast to the situation for radiological land contamination, little or no work has been done in the area of ecological consequences of hazardous chemical contamination. Therefore, discussion of ecological consequences will be limited to radioactive contamination and consequences of nuclear war.

## 3.4.1 Direct and Indirect Radiation Effects

The range of radiosensitivity among plant species varies by several orders of magnitude. Genetic effects have been demonstrated among sensitive species exposed to natural terrestrial radiation as low as 0.1 mrad/d (Mericle and Mericle 1965). Among woody species, conifers (pine, spruce, fir, etc.) are among the most radiosensitive species (Sparrow et al. 1963b). The order of decreasing radiosensitivity is generally considered trees > shrubs > herbs > thallophytes (algae, fungi, lichens). Table 3.7 lists relative radiosensitivities of some economically important animals and plants. The values in Table 3.7

Table 3.7. Relative radiosensitivities of economically important livestock and plants  $\underline{\underline{a}}$ 

Item	LD50 <u>b</u> (x1000 R)	Item (x1000 R)	LD <sub>50</sub>
Cattle	0.5	Corn	>15
Sheep	0.4	Oats	17-27
Swine	0.6	Wheat	14-25
Chicken	0.9	Lettuce	5
Cabbage	<b>-</b> 11		
Pine	0.7	Pepper	24
0ak	4	Tomato	13-37
Hickory	4-6	Potato	1.6-10
Fescue grass	3	Onion	1.5
White clover	11-24	Beet	1.6

aSources are Sparrow et al. (1971) and Bell (1971).

 $\underline{b} \text{LD50}$  is the radiation dose which is lethal to 50% of the exposed individuals.

should be used as a rough guide to radiosensitivities; under actual fallout conditions greater or less sensitivities would likely be encountered.

Radiation effects on terrestrial ecosystems are termed direct or indirect effects. Direct effects include increased mutations, reduction in reproductive capacity, and somatic or physiological responses resulting in growth inhibition or mortality. Indirect effects within natural plant communities include changes in form and structure, species composition, and species diversity, as well as in productivity. These may result from alterations in spatial relationships, competition, nutrient availability, and light and moisture relations. In general the principal effects of ionizing radiation on natural terrestrial ecosystems are (Miller and LaRiviere 1966):

"... the simplification of the ecosystem by selective mortality or growth inhibition of sensitive species. These changes in plant populations would be expected to cause changes in insect populations since the latter would be expected to be sensitive to the abundance of food supplies. In these damaged ecosystems, the capacity of the ecosystem to recover should remain intact, at least initially, but rapid changes in plant species composition and in number of plants during the first few years after injury would be expected to occur. Reduction in competition, appropriate radiation exposures, and other factors would result in stimulated growth patterns of some species and retarded growth for others..."

One of the most recent assessments of the effects of nuclear contamination was a committee report commissioned by the National Academy of Sciences to consider the long-term worldwide effects of multiple nuclear-weapons detonations (NAS 1975a). That assessment was limited to the effects within 30 years on natural terrestrial ecosystems remote from nuclear detonations in the northern hemisphere.

From a total yield of 10,000 megatons from multiple detonations, the committee concluded that the accumulated average total exposure would be 2 rads above background, and that there would be no widespread ionizing radiation effects on the plant components of natural terrestrial ecosystems during the period one week to 30 years after detonations.

However, localized areas adjacent to detonations could experience exposures 20 times greater than average (United Nations 1972). The average total exposure at 30 years would be approximately 100 rads. Slight growth inhibition among radiosensitive species would occur in some ecosystems or communities (McCormick 1969). The structure of these ecosystems would not be significantly altered, and recovery would be complete by 30 years postdetonation. The best available and most appropriate descriptions of ecosystem recovery capacity are found in several studies following acute ionizing radiation exposures of several hundred times current levels. In these studies, recovery follows normal predictable successional patterns, except that recovery often has been faster than predicted (Whicker and Fraley 1974).

If the radiation doses were greater, then ecosystems or plant communities dominated by radiosensitive species would experience physiological and genetic damage, increased mortality, and decreased production (Rudolph 1971). The coniferous forests of the north temperate zone would be especially vulnerable.

The timing of the fallout would be important. Fallout early in the period of active growth would enhance the response of some species. Conifers irradiated during the period of active growth are

more radiosensitive than when irradiated during periods of dormancy (Sparrow et al. 1963b; Taylor 1966). Less sensitive species might experience slight to severe growth inhibition, depending on their relative radiosensitivity. Nonconiferous species are generally less sensitive to ionizing radiation than conifers with the order of decreasing sensitivity being angiosperm trees > shrubs > thallophytes (lichens, fungi, etc.) > microflora. Thus, there would be an initial change in the direction of decreased diversity and increased importance of shrubs, "weedy" species, and lower forms.

Dormant, viable seeds and subterranean roots are afforded a degree of radiation protection, either by being physiologically inactive or by shielding effects of substrata (soil). The recovery of plants within a damaged ecosystem would depend on the availability of these propagules, and reestablishment of a dominant plant community would depend on the edaphic (soil) and climatic conditions. Recovery would be expected to follow ordinary successional processes similar to recovery from other stresses such as fire, herbicides, or mechanical disturbance (Osburn 1968). However, in areas where ionizing radiation eliminates most or all flora, soil erosion and leaching of nutrients could severely hinder recovery.

#### 3.4.2 Increased Ultraviolet Radiation

According to the committee to study the long-term worldwide effects of nuclear weapons detonations, there would be a significant increase in biologically active ultraviolet (UV-B) solar radiation (290-320 nm wavelength) exposures to natural ecosystems following

multiple nuclear-weapons detonations of 10,000 megaton yield due to ozone depletion (NAS 1975a). During the first year following detonations, uninvolved northern hemisphere nations could experience UV-B increases as great as 650%. Southern hemisphere nations would receive significantly lower UV-B exposures than would northern hemisphere nations in close proximity to the detonations. Global increases during this time might range from 20 to 120%. After three years, UV-B flux would tend toward equilibrium at a level approximately 9% above current levels.

A great variety of species respond to changing intensities of UV-B. A report by the U.S. Department of Transportation (1974) concludes that for man there is a relationship between assumed UV-B exposure and the incidence of skin cancer. In the event of multiple nuclear-weapons detonations, severe disabling sunburn could result from an exposure to the sun for 1 h. Additional studies, although inconclusive, have demonstrated that UV-B exposure inhibits plant growth and development, increases mutation rates of accessory reproductive cells, reduces photosynthesis, and influences the pollination behavior of insect species. Further evidence indicates that the current UV-B flux is sufficient to kill many species and that others survive at an upper limit of tolerance. For these organisms, any increase in UV-B flux, or any disruption of their protective habitat, could threaten survival.

## 3.4.3 Local Climatic Changes

In the event of a nuclear war, dramatic changes in microclimate would be expected in areas devastated and denuded by nuclear detonations or extensive release of toxic chemicals. For example, at Copperhill,

Tennessee, toxic sulfur dioxide fumes released in the late nineteenth century devegetated an area of approximately 28 km<sup>2</sup> (7000 acres) and affected an additional 100 km<sup>2</sup> (25,000 acres), causing extensive erosion of top soil. As a result, air and soil temperatures, evaporation, and wind velocity increased; rainfall and soil moisture decreased; and greater temporal variation in climatic conditions occurred (Ingersoll 1964).

Of the climatic parameters, fluctuations in both the means and the extremes would be of interest from the standpoint of ecological effects. However, the alterations, if any, in temperature and precipitation regimes would probably be temporary unless permanent loss of ecosystem structure occurred as in the case of Copperhill, Tennessee.

## 3.4.4 Nuclear Winter

In 1975 it was estimated that dust particles would enter the earth's atmosphere following 10,000 megatons of nuclear detonations (NAS 1975a). Most of this dust would be removed from the atmosphere by precipitation during the first year after injection, but would result in a worldwide reduction in incident solar radiation at the earth's surface sufficient to reduce the mean annual temperature by perhaps as much as 0.5°C and light intensity (estimated from the known effects of volcanic dust) by 2-3% (Mitchell 1973). Conditions attendant on the depletion of ozone and increased UV-B radiation might perhaps result in an additional 0.5°C temperature decrease. A 1°C global average temperature depression was thought to be the most extreme that might occur, and it was felt that a considerably smaller temperature lowering would be more realistic.

However, a more recent investigation of the potential global atmospheric and climatic consequences of a nuclear war postulates significant attenuation of the solar radiation flux and subfreezing land temperatures resulting from fine dust raised in high-yield nuclear surface bursts and from smoke from city and forest fires ignited by airbursts (Turco et al. 1983). This recent study is generally referred to as the "TTAPS study," after the last names of the authors (Turco, Toon, Ackerman, Pollack, and Sagan). In the TTAPS study, the dust and smoke generated from the exchange of 5000 megatons would encircle the earth within one to two weeks, thereby reducing light intensities over a period of many weeks, with land temperatures reaching -15 to -25°C. Such subfreezing land temperatures could persist for months, even if the nuclear exchange came during the summer. With perturbations to global circulation patterns, and changes in precipitation, nuclear war survivors might experience, in any season, what is known popularly as "nuclear winter". A nuclear winter in the northern hemisphere might reduce biological productivity for a year or more, with war survivors subjected to starvation in addition to freezing temperatures and exposure to ionizing radiation (Ehrlich et al. 1983).

The results of a more recent study commissioned by the National Research Council were generally consistent with the findings of the TTAPS study (National Research Council 1985). In the council's study, the scenario is based on a nuclear exchange of approximately one-half of the estimated total world arsenal of nuclear weapons (6500 megatons). The council's conclusions differed from the TTAPS study in that the postulated nuclear winter might not be as severe as the latter

concluded. For example, an attack during the summer might decrease the mean continental temperature in the northern temperate zone by 10 to 25°C, whereas an attack of the same magnitude during winter might produce little change in the mean continental temperature. The Council's study also concentrated on the northern hemisphere, but also speculated on possible effects in the southern hemisphere. While southern hemisphere effects would be less extensive, significant quantities of dust and smoke could be transported across the equator as the result of a heating-enhanced circulation in the debris cloud.

In any nuclear exchange of the magnitudes assumed in the two studies (Turco et al. 1983; National Research Council 1985), global atmospheric and climatic consequences (nuclear winter) might be sufficient to cause extinction of a major fraction of plant and animal life, with serious implications for the survival of man.

#### 4. COUNTERMEASURES

# 4.1 RADIOLOGICAL STANDARDS

In the event of a land contamination event involving radioactivity, it is not clear what federal radiation standards may apply, or whether any federal radiation standards will apply (in the event of a nuclear war existing standards will undoubtedly be revised upward). In general, radiation standards are designed to address the licensing, construction, operation, and decommissioning of various industries which mine, manufacture, or utilize radioactive materials. There are no standards written specifically for the cleanup of land contaminated via a nuclear detonation, large reactor accidents, broken arrows, or other unspecified accidents involving radioactive materials. However, standards written for protection of the general public and employees in nuclear fuel cycle occupations have been used in the past as guidelines for cleanup of contaminated lands (DOE 1982). Therefore, it is expected that in the future these standards will also be used as guidelines.

The Nuclear Regulatory Commission (NRC) has established radiation protection standards for occupational exposures arising from activities under licenses it issues in Title 10 of the <u>Code of Federal Regulations</u>, Part 20 (10 CFR 20). In 10 CFR 20, the limit of exposure in restricted areas is 5 rems/y for whole body dose. For unrestricted areas and for minors (under age 18) in restricted areas, the limit is 500 mrems/y. Since these are occupational exposures, they may be interpreted broadly as maximum doses.

The EPA has established radiation protection standards for the general public for nuclear fuel cycle operations (40 CFR 190) and for residual radioactive material at uranium milling sites (40 CFR 192). Under 40 CFR 190, annual doses to the general public are limited to 25 mrems to the whole body, 75 mrems to the thyroid (from iodine-129), and 25 mrems to any other organ. Under 40 CFR 192, limits are imposed on surface soil concentrations of radium-226 in contaminated areas exceeding 100 m<sup>2</sup> and on air concentrations of radon-222 and daughters.

As stated before, the federal radiation protection standards are designed specifically for licensing and operation of nuclear facilities. In the event of land contamination that requires remedial action, it is not clear that any standards can be applied in a legal, binding sense. Instead, radiation protection standards are likely to be used as guidance as to what doses are considered to be "safe" or "acceptable." Under 10 CFR 50 (Domestic Licensing of Production and Utilization Facilities), the NRC specifies that effluents be kept to "as low as reasonably achievable." This approach appears to be more robust than the establishment of an absolute limit. As will be seen in the following sections, "as low as reasonably achievable" may be based on such considerations as cost, decontamination efficiency, and other factors.

#### 4.2 FOOD MONITORING AND CONTROL

After land is contaminated, protection against external radiation doses or direct exposures to toxic chemicals may be accomplished through radiation shielding and/or denial of access to the area.

However, protection against doses of lower intensity, internal doses, and long-term effects from chronic exposures would require monitoring and sheltering to control external exposures and alteration of agricultural practices to control ingestion (internal) exposures from foods produced on the contaminated land. Gant and Chester (1981) estimate that an effective civil defense program which limited or controlled exposures after a nuclear war could significantly reduce radiogenic cancer deaths. They estimated an average loss of life expectancy for war survivors with good shelter could be kept to 1.2 years or less (and 0.2 year by controlling exposures of the population under age 40).

The survival of food crops and livestock was the subject of a symposium held in 1970 (Bensen and Sparrow 1971). Radiation effects, properties of fallout, radiation pathology, measurement techniques, and implications for civil defense planning were subjects covered in the four-day symposium. The reader is referred to Bensen and Sparrow (1971) for more detailed information on the subjects covered in Sect. 4.2.

Protection of livestock from ionizing radiation may be accomplished through shielding (sheltering), preventing access to contaminated forage, and skin decontamination (Byrne and Bell 1973). In the event of a nuclear detonation, protection is most effective during the first 24-48 h from the start of fallout (Olsen 1965). If livestock are not sheltered before contamination of the land via fallout, they will be exposed to radiation from external contamination and to internal contamination from ingested feeds and inhaled particles

[(radiation effects, survival rates, and salvage/utilization of livestock are reviewed thoroughly by Byrne and Bell (1973)]. However, if exposed animals are placed on a diet of uncontaminated feed and are placed in a sheltered environment, contamination levels in milk and eggs will reach normal levels within one week because of the fairly rapid turnover times for these two products (Ng et al. 1977; Ng et al. 1982). Turnover rates for tissues such as muscle, liver, kidney, and other internal organs, however, are much slower, and decontamination through radioactive decay (i.e., storage of dried food) may be necessary.

Pathological effects of ionizing radiation on livestock can range from death at high exposures to burns from beta radiation to loss of appetite and weight. The most severe damage from ingestion of contaminated feed by livestock is to their gastrointestinal system (Byrne and Bell 1973). Whole body gamma irradiation generally results in variable injury to different tissues, with radiosensitivity generally lymphoid tissue > bone marrow > spleen > gonads > gastrointestinal tract (Olsen 1965). Depression of normal immunological mechanisms in animals exposed to gamma radiation could result in increased incidence of bacterial infection, and make them unfit for human consumption. Because several days may elapse between exposure and symptoms of radiation sickness, it is suggested that slaughtering during the symptom-free period be done only after ascertaining that the animal's body temperature is normal (Olsen 1965).

Reduction of the amount of contamination ingested by livestock can be achieved through reduction in feed allotments, fasting for several days, or use of stored grains and protected water, e.g. groundwater, which is less susceptible to contamination due to low flow rates (Olsen 1965). Generally speaking, plant uptake of radionuclides is ordered fine roots > leaves and stems > fruits, seed, and tubers (Baes et al. 1984), so use of grains, concentrates, or stored silages would be preferable to fresh silages and hay and unconfined grazing of pasture where fine roots and soil may be ingested.

For protection against iodine-131-contaminated milk, delayed marketing is the key because of iodine-131's short half-life of eight days. Significant protective measures include removal of lactating cows from pasture and use of stored feeds; withholding and diversion of milk to manufactured products to allow for radioactive decay; storage of fresh concentrated milk, frozen fresh milk, and concentrated milk; and use of ion exchange resins for removal of iodine-131 (Todd 1965).

Reduction of radionuclide contamination in plant foods for human consumption can be accomplished through altered agricultural practices, food storage, and food preparation and processing (Todd 1965). The critical radionuclides from the dietary contamination viewpoint are iodine-131, strontium-89, strontium-90, cesium-134, and cesium-137, however after six weeks the importance of strontium-89 is significantly diminished (Russell 1965). For the short term, only surface contamination of vegetable crops and iodine-131 contamination of milk are of importance. Surface contamination of leafy and vegetable crops can be greatly reduced by preparation (e.g., removal of outer leaves of lettuce and cabbage) or peeling. For many vegetables and fruits, outer tissue layers are normally removed prior to cooking and/or consumption. These produce include citrus fruits, corn, peas, many

beans, melons, and nuts. For fruits and vegetables with hairy surfaces or irregular shapes (e.g., peaches, asparagus, broccoli) there may be no satisfactory method of decontamination (Todd 1965), and consumption of contaminated parts should be avoided.

Root uptake of radionuclides deposited in soil via fallout is of greatest long-term importance. If fallout contaminated land must be used for the production of food and feed crops, then several measures can be taken to reduce the amount of contamination taken up by the crops. First, existing crops and grasses can be removed along with the surface soil layer. Of course, costs will be quite high. Furthermore, such measures are only practicable if the contaminated area is relatively small. Additional measures to reduce the uptake of strontium and cesium into agricultural plants include:

- Deep plowing of the soil to lower contamination concentrations in the upper soil layers and reduce uptake by shallow-rooted plants (e.g., grasses and legumes)
- Addition of phosphorous to soil to reduce plant uptake of strontium via the formation of insoluble strontium phosphates
- Addition of calcium (lime) to soil to reduce plant uptake of strontium by increasing the calcium/strontium ratio of the soil [However, excessive calcium, magnesium, or stable strontium additions can actually enhance strontium uptake, depending on soil pH and cation exchange capacity (Francis 1978)]
- Addition of potassium (potash) to soil to reduce plant uptake of cesium by increasing the potassium/cesium ratio of the soil
- Using deep-rooted plants (e.g., alfalfa and corn)
- Addition of chelators or solvents to move the contamination below the root zone (not recommended in areas where contamination of groundwater supplies is a potential problem)
- Choosing plants with high calories-to-calcium ratios (e.g., cereals, sugar crops) or fiber-producing plants (Comar 1962)

## 4.3 LAND DECONTAMINATION

Land decontamination involves removal, degradation, or isolation of the contaminating material such that it is no longer a threat to human health and safety. For decontamination of structures affected by radioactive fallout or toxic chemicals, methods include firehosing or flushing with water, application of a fixative, sandblasting, vacuuming, and packaging and removal. For nonpaved land areas, methods include scraping, grading, vacuuming, bulldozing, removal of vegetation, and covering with soil, asphalt, or sewage sludge (Miller 1963; Smith and Lambert 1978). During decontamination operations, analytical or radiological support will be necessary, as will a method for "ultimate" disposal. In some instances restriction of land use and access will be necessary. However, land restriction is but a temporary solution. At some time, provisions for unrestricted land use will be necessary.

### 4.3.1 Natural Removal Mechanisms

Once land has become contaminated various natural mechanisms will begin to both remove the material and enhance its dispersion. Such mechanisms include radioactive decay; chemical, biological, or photo degradation; leaching; resuspension; and animal vectors. Decay and degradation of the contaminant will remove the material from the land area; the other processes will both remove and disperse the material from a given area.

4.3.1.1 Decay and Degradation. Typically, radioactive decay and degradation of chemicals in the environment are measured in terms of "half-life" or "half-time" that describe the amount of time that it

takes for one half of the material to physically disappear. Four half-lives will result in less than 10% of the material remaining; seven half-lives will result in less than 1% of the material remaining. Thus, contamination with materials with half-lives or half-times of less than a few years require only temporary control and restriction of the contaminated land area. Contamination of land with materials with half-lives and half-times of tens to hundreds of years requires a more "permanent" solution. For radionuclides, half-lives are constant and well known (Kocher 1981). For toxic chemicals, half-times are variable depending on environmental factors. Persistence information has been tabulated for some pesticides (Trabalka and Garten 1982), but generally, environmental half-times are unknown. Even when persistence studies are performed, results can show great variability. For example, based on early U.S. Air Force soil tests, it was thought that dioxin's environmental half time is about 250 days (Hay 1976c); however, studies of dioxin in Seveso, Italy, soils indicate a half-time of perhaps ten years or more (Kriebel 1981).

4.3.1.2 Leaching and Runoff. Material deposited on land surfaces will initially be retained there, but it may later be redistributed through the action of precipitation. The material may be leached downward through the soil profile or moved laterally (or downslope) via surface runoff. Leaching and runoff may initially reduce the availability of the material for direct contact with man and agricultural products at the location of initial contamination, but these processes may also be responsible for enhancing contaminant dispersal. Chemical treatments to enhance leaching have been suggested

as a mitigation technique for radionuclides deposited in surface soils (Comar 1962); however, the potential for contamination of ground or surface waters would argue against the approach, especially for long-lived radionuclides. Currently, nearly half of the U.S. population use groundwater from wells or springs as their primary source of drinking water. Thirty-six percent of municipal public drinking water comes from groundwater sources, and 75% of the major cities in the United States depend on groundwater for most of their supply (Pye and Patrick 1983).

4.3.1.3 Resuspension and Animal Vectors. Material deposited on the surfaces of soils and vegetation may be moved through the action of wind and biological vectors. For example, resuspension of radioactive tailings from mining and milling activities in the western United States is a major source of population exposure to decay-chain daughters of uranium radionuclides (EPA 1979). Linsley (1978) has reviewed resuspension as a mechanism for transfer of transuranium elements and models for quantifying resuspension. In general, resuspension of particulate matter deposited on surfaces is greatest immediately after deposition, with an initial rapid decline in air concentration, followed by a more gradual decline. Moisture is an important factor in reducing resuspension, as is the effect of soil cover (vegetation). Human activities, in addition to wind-driven processes, may be responsible for resuspension of particles. Some examples are pedestrian or vehicular traffic, digging, and plowing. Linsley (1978) points out that most information on resuspension is derived from measurements of the relationship between surface and

airborne contamination following nuclear weapons testing, and most information pertains to semi-arid environments. Such data may not be applicable to (too conservative for) land areas with greater vegetation cover and moisture regimes.

Contaminants may also be dispersed under the influence of animal vectors. For example, Cadwell and Fitzner (1984) describe the occurrence of radionuclides in feces from birds of prey (raptors) nesting near contaminated lands. The activity in the feces (castings) reflects the intrusion of burrowing rodents and reptiles into contaminated soils, radionuclide uptake, and the transfer from prey to predator. Burrowing mammals, rodents, reptiles, arthropods, and annelids can invade buried contamination, ingest it, and provide a vector for its transport to uncontaminated areas. Grazing herbivores, likewise, can ingest surface contaminated vegetation and soil and disperse the contamination. Predators feeding on these vectors can both bioaccumulate the contamination and disperse it over large range areas. In the Cadwell and Fitzner (1984) study, nesting sites were occupied over many years and proved to be highly contaminated. Such biological transport is rarely accounted for in contaminated land management plans.

## 4.3.2 Options for Achieving Land Decontamination

Management of contaminated lands may be based on social and political issues and consequences as well as scientific concerns.

Definition of when the area is "safe" for human habitation or unrestricted use may have to be based on the former rather than on the latter. Performance of a risk assessment which finds no significant

health impact may not be enough to convince residents affected by the land contamination event to remain in (or return to) the area. Historical evidence from past contamination events is that remedial actions are largely based on "being safe, rather than sorry" instead of on health risk assessments or acceptable government standards. When the contamination event occurs, public pressure to "do something" increases. Public perception that remedial actions are being initiated by local, state, or federal officials often does much more to alleviate this pressure than official acclamations that health risks are insignificant.

Therefore, it is essential in management of contaminated land areas that the public perceives that the problem is being actively addressed. Here "active" means that procedures are implemented to contain the contamination, limit human exposures, and decontaminate or detoxify the affected areas. Waste containment and management might include (Wright and Caretsky 1980):

Runoff control

including drainage ditches, channels, subsurface drains, dikes, and culverts to divert water from the area or contain it to prevent extensive off-site (downslope) dispersion during runoff. including caps, surface seals, and subsurface walls to prevent extensive lateral dispersion of the contaminant or eliminate infiltration of water and leaching.

Barriers

Stabilization

with vegetation or well-compacted, fine-grained soil covers to reduce runoff velocities (and erosion) and minimize infiltration.

Monitoring

including air, soil, surface water, and groundwater monitoring to detect movement of the material away from the initial contamination area. Monitoring is also necessary to define the physical extent of the contamination, the effectiveness of decontamination procedures, and when the problem no longer exists.

Security

to prevent unauthorized entry into the site.

Treatment

including in situ biological, chemical, and physical treatment techniques to degrade or detoxify contaminants or fix them in place.

Recordkeeping

including waste characterization, monitoring data,

inspection results, and maintenance performed.

Risk assessment

to evaluate in a quantitative manner the exposures, population-at-risk, health risks, and uncertainties

Before cleanup techniques and decontamination procedures are implemented, decisions must be made regarding priorities, costs, and extent of cleanup. Regarding cleanup priorities, Unterberg et al. (1982) suggest the following order: human populations and habitats > fauna, > flora, > properties, > and aesthetic and recreational areas. Finally, they recommend use of judgment and common sense in setting these priorities. Once a set of cleanup alternatives for a site has been established, factors such as available equipment, manpower, and financial resources will limit the response. Cost may be the overriding factor, but such factors as completeness and rate of

associated with the contaminants.

contaminant cleanup, logistics, environmental impacts, personnel protection, and "ultimate" disposal problems will be important.

Often it is not clear just how clean is "clean." The extent of cleanup can be based on government standards (acceptable contamination levels), the change in effectiveness of cleanup as contamination levels decrease, change in incremental cleanup costs as levels decrease, environmental impact of the cleanup techniques, public awareness and political pressures, budget, and time constraints. There appear to be no universal rules. Each site must be evaluated individually.

Costs of cleanup will invariably be high. Moein (1978) estimated the costs of cleanup of various types of hazardous substances as follows:

- \$2.20/kg (\$1/lb) of soluble substance on land (e.g., acid spill from tanker trailer)
- \$1.85 per liter (\$7/gal) of floating or oil-like substances in water
- \$53 to \$320 per liter (\$200 to \$1,200/gal) of insoluble substance on land or sinking substance on water
- \$160 per liter (\$600/gal) for PCB spills

The estimated cost of cleanup of a 14-ha (35-acre) tract on Neville Island, Ohio, which had been a waste disposal site for benzene, phenols, parathion, coal tar residues, and trace metals, was estimated to be from \$7 to \$24 million (Brunwasser and Spence 1980). Smith and Lambert (1978) discussed the technology and costs for cleanup of land contaminated with plutonium. Their results are broadly applicable for other long-lived radionuclides and toxic metals. Their results are given in Table 4.1.

Table 4.1. Costs associated with cleanup of contaminated landa

Method	Average cost (dollars per hectare)	Range (dollars per hectare)
Fencing off land	2,700	860-8,200
Chemical or vegetative land stabilization	5,200	1,200-7,400
Deep plowing	6,400	2,200-12,000
Applying 10 cm (4 in) soil cover	12,000	5,900-19,000
Applying 30 cm (12 in) soil cover	24,000	13,000-35,000
Soil removal/scrape into windrows	8,900	4,000-16,000
Soil removal/scrape into mounds or trenches	13,000	5,400-25,000
Soil removal/offsite disposa (non-retrievable)	310,000	27,000-470,000
Soil removal/offsite disposa (retrievable)	1 410,000	370,000-5,800
Soil removal/offsite disposa (federal repository)	1 1,300,000	990,000-1,500,000

≜After Smith and Lambert (1978).

### 4.4 ALTERNATIVE LAND USES

After implementation of remedial action, or in the event that land decontamination alternatives are impractical, the question of the ultimate use of the contaminated land still remains. The most important consideration is still protection of the public from health hazards, but if possible, it would be advantageous to derive some economic benefit from the land.

If the contaminated land was formerly developed, it seems prudent to raze buildings and structures to eliminate the potential intrusion by looters and the curious. In such instances the construction of parking lots, parking garages, or other facilities requiring a minimum of attention may be possible.

In the case of agricultural land, crop selection can minimize potential exposures to contaminants. Use of grain or seed-stock producing crops would take advantage of the generally lower concentration factors for reproductive plant parts. Use of fiber, sugar, or oil-producing crops would take advantage of the decontamination factors afforded by the processing required to produce these products. At least a switch from use of leafy vegetables with large surface areas to crops for which the edible part is somewhat protected from atmospherically deposited material would be beneficial.

If the contamination of agricultural land were so severe as to preclude active agricultural management practices, then a passive approach of allowing for natural forest succession or aerial seeding is an attractive alternative. Economic benefit (timber production) might

be derived from the land while radiological decay or environmental degradation of the contaminant occurs. Secondary succession (succession from an area in which the vegetation community was destroyed or removed) to a mature pine forest takes between 25 and 100 years (Odum 1971). However, severe contamination of the most environmentally persistent contaminants could preclude commercial use of timber harvested from the area.

Reforestation would be compatible with fencing off the area or other security measures. Closing of the forest canopy and establishment of an organic litter layer on the forest floor would also tend to fix the contaminant in place (thereby reducing leaching and biological uptake), provide a warm, moist medium for microbial and fungal populations (thereby enhancing biological degradation), and reduce wind (resuspension) and rain action (erosion).

## 5. CONCLUSIONS AND RESEARCH NEEDS

## 5.1 CONCLUSIONS

An extensive review of Civil Defense reports and the open literature revealed that sources, impacts, and countermeasures for land contaminated by radionuclides are well documented, and there are relatively few sources of radionuclide that are of concern for long-term land contamination.

Radionuclides of concern from nuclear weapons detonation are tritium, carbon-14, cobalt-60, strontium-90, ruthenium-106, iodine-129, cesium-134, cesium-137, cerium-144, plutonium-238, plutonium-239, and americium-241, but only strontium-90, cesium-137, iodine-129, and the transuranics are of significant importance from a long-term land contamination standpoint. For a reactor accident, most of the above nuclides may be released and also plutonium-240, plutonium-241, curium-242, and curium-244. In a serious reactor accident only strontium-90, cesium-137, and the transuranic radionuclides are of long-term importance.

Reactor accidents have not produced the levels of contamination hypothesized in reactor accident risk studies. With the exception of the Three Mile Island (TMI) accident, the serious events have been in experimental or prototype reactors. None of these reactor accidents have been harmful to public health. For the TMI accident, the maximum estimated radiation dose received by any individual in the off-site population was about 70 mrems. The collective dose to the surrounding population within 50 miles was about 0.8% of annual background doses from natural radiation sources. These doses were mainly due to the biologically inert noble gases which presented no land contamination problem.

Since the end of World War II there may have been approximately 200 accidents involving nuclear weapons ("broken arrows"), including weapons released in airplane crashes, missle explosions, and nuclear materials released in fires. So far, such accidents have not led to nuclear explosions because of numerous safety design features. Impacts of broken arrows have included millions of dollars in property losses and cleanup costs and dispersal of kilogram quantities of uranium and plutonium over areas ranging from a few square meters to several hundred hectares. Food-chain transport of plutonium and uranium are of minor concern; the predominant exposure pathway is from inhalation of resuspended material or inadvertent handling of nuclear fuel debris. In general, cleanup consists of the removal of debris and contaminated soil. According to the U.S. Air Force, nuclear weapons are no longer being carried routinely on training missions, and, therefore, the frequency of broken arrows should be far less than in the past.

The effects of ionizing radiation on man, livestock, and natural ecosystems have been extensively studied, and only a few radionuclides (e.g., strontium-90 and cesium-137) present long-term land contamination problems. Biological effects of ionizing radiation, ranging from radiation sickness to death, are associated with doses to man that exceed 100 rems over a short period of time. The health effects expected from exposures to low levels of radiation include small increases in the number of cancers among exposed populations and potential genetic effects that may be expressed in future generations. However, both health and genetic effects are predicted by linear extrapolation from higher dose levels. Such effects from low doses

have not been directly observed in man (e.g., the survivors of the Hiroshima and Nagasaki weapons blasts).

Sources, impact, and countermeasures for land contaminated by hazardous chemicals are not as clearly defined, documented, or understood as for radionuclides. For the nearly 200,000 chemicals of commercial significance, perhaps as many as 30,000 are hazardous. They can be toxins, mutagens, teratogens, or carcinogens and also present corrosion, fire, or explosion hazards. Their entry into the environment can occur in many ways, including improper handling, transportation accidents, and industrial accidents. While most accidents involving radionuclides have been transportation related, most serious hazardous chemical releases have been related to illegal or improper waste management practices. Information on a chemical's environmental behavior, persistence, and toxicological properties is often lacking, and analytical standards, laboratory methods, and quality control procedures are often unavailable.

In general, indirect modes of exposure to toxic substances (food-chain transport) are of secondary importance to direct modes of exposure (direct contact, inhalation) because food-chain transport involves transfers across one or more biological membranes where the potential for discrimination against contamination exists. However, there are exceptions to this generalization, especially when short cuts in food-chain transfer bypass these barriers or when the compound's chemical properties result in its bioaccumulation or bioconcentration in plants or animals.

The biological behavior of radionuclides and toxic metals has been extensively studied, and compilations of transfer factors for plants and animals are available. The environmental behavior of organic compounds, however, is very difficult to predict with accuracy. Some predictions can be made on the basis of water solubility, octanol-water partitioning, and other measures, but confidence in such models is low. Some of these generalizations are the following:

- Environmental conditions or biological processes that increase the water solubility of a contaminant will tend to increase its entry into vegetation via plant roots.
- The uptake of radionuclides and organic chemicals is greatest in sandy soils having low clay content and low organic matter.
- Volatilization of organic chemicals from soil and recondensation on plant surfaces can be a far more important contamination pathway than uptake from soil via plant roots.
- Uptake and apportionment among roots, stems, leaves, and edible parts will vary with the contaminant studied, its metabolism in the plant, plant age, and plant species.
- In the absence of significant root uptake and translocation, it is still possible to have accumulation of contaminants in root crops because of surface adsorption.
- The persistence of an organic chemical in soil will govern its potential for uptake by plant roots and subsequent transfer through the food chain.

Generalizations regarding entry of inorganic and organic contaminants into plants through direct deposition on plant surfaces can also be made:

 Direct deposition is most important for food crops such as lettuce, cabbage, and other leafy vegetables. For these crops, plant parts exposed to direct deposition are consumed.

- Deposits of large particles on vegetation will be rapidly lost through exposure to natural meteorological processes, but submicron particles may be retained indefinitely, depending upon the properties of the leaf surface.
- There are no adequate simple models for predicting absorption of volatile organics by foliage.

There are several important generalizations regarding food-chain transport of contaminants in animals and in assessing the use of contaminated agricultural lands.

- Inorganic and organic substances differ in their metabolism, tissue distribution, and excretion from the body. This fact makes it very difficult to predict food-chain bioaccumulation of complex mixtures of contaminants without specific information on the animal species and the particular contaminants involved.
- Ingestion of contaminated soil can be more important or just as important to the food-chain transfer of contaminants as ingestion of contaminated forages.
- Organic substances of most concern for bioaccumulation in terrestrial animals are lipophilic compounds that are environmentally persistent. Such compounds (e.g., polycyclic aromatic hydrocarbons) have environmental half-times in soil greater than 14 days and octanol-water partition coefficients greater than 3000.
- Agricultural practices which increase the mobility of contaminants in soil may also increase their potential uptake by livestock.

Protection of foods following a large radiological land contamination event may require some alteration of agricultural practices. Food producing animals can be sheltered, placed on stored feeds, and fasted to reduce contamination levels in their food products. Once exposed to a diet of uncontaminated feed in a protected environment, contamination levels in milk and eggs will reach normal levels within about one week. Clearance times for liver, kidney, and

other internal organs are much longer. Consumption of these organs would not be recommended. Depression of normal immunological mechanisms in animals exposed to gamma radiation could increase incidence of bacterial infection, making them unfit for human consumption, unless slaughtering takes place during the symptom-free period when body temperature is normal. Delayed marketing of milk contaminated with iodine-131 is a key protective measure because of its short eight-day half-life.

Reduction of radionuclide contamination in plant foods for human consumption can be accomplished in several ways, including food storage, food preparation, and processing. Surface contamination of leafy vegetable crops can be reduced by removal of outer leaves, washing, or peeling. For fruits and vegetables with hairy surfaces or irregular shapes (e.g., asparagus, broccoli) there may be no satisfactory method of decontamination. Peeling and discarding exposed parts or avoidance is recommended for these types of fruits and vegetables. Additional measures to reduce radionuclide uptake into agricultural plants include:

- deep plowing of the soil to lower contamination concentrations in the upper soil layers and reduce uptake by shallow-rooted plants,
- addition of phosphate, lime, and potash to reduce strontium and cesium uptake,
- using deep-rooted plants,
- addition of chelators or solvents to move the contamination below the root zone in areas where contamination of water wells is not a potential problem, and
- utilizing plants with high calories-to-calcium ratios or fiber-producing plants.

Planning for reclamation of land contaminated via nuclear war will involve different assumptions and criteria for reclamation than for smaller events such as reactor accidents or weapons accidents. In a small radiological contamination event it is likely that cleanup will be undertaken, despite the high cost. After a nuclear war, however, total cleanup will likely be impossible and mitigation measures such as deep plowing and liming of agricultural soils, removal of lactating cattle from contaminated pasture, use of stored feeds, and food processing and storage to reduce exposures likely will be instituted. Furthermore, standards for cleanup will likely be much more liberal than current standards, which limit both occupational and nonoccupational exposures in the nuclear industry.

Remedial actions and land decontamination alternatives are few and very expensive. The most extensively used option is removal and burial (or destruction) of contaminated materials. When removal and disposal of contaminated vegetation, structures, and soils have not been practicable, isolation of the land from public use has generally been the approach taken. In the case of agricultural land, conversion to pasture or rangeland, production of fiber or seed-stock crops, production of biomass for fuels, or allowing for natural forest succession or planting of tree seedlings for timber production are possible alternative land uses which derive economic benefit from the land. Timber production allows the land to be secured from human intrusion (fenced off) while radiological decay or environmental degradation of the contaminant occurs. For hazardous chemicals, more options exist for land decontamination than for either radionuclides or

toxic metals, including biological, thermal, or photodegradation of the contaminant. However, most of these methods are based on laboratory studies, are unproven in the field, and are more expensive than traditional methods.

Time and nature will both aid and hinder the land decontamination process by removing the material and enhancing its dispersion. The mechanisms which remove the contaminant include radioactive decay and chemical, biological, or photodegradation. Leaching, resuspension, and animal vectors will tend to disperse the material. Radiological decay and environmental degradation are measured in terms of "half-life" or "half-time," respectively. Half-lives of radionuclides are known with certainty, but for organic chemicals, persistence information is sparse. The action of wind and precipitation can result in leaching, runoff, and resuspension of a contaminant. Burrowing animals may physically move the contaminant from one location to another. This dispersion may slightly reduce its availability at the site of initial contamination, but may present additional problems at other locations in other media (e.g., groundwater contamination). This is one reason why chemical treatments to enhance leaching below the root-zone of agricultural plants are not recommended here (chemical treatments are also very expensive).

Decontamination and remedial action procedures commonly used in the control and management of contaminated lands include:

 runoff control through the use of drainage ditches, channels, subsurface drains, dikes, and culverts to divert water from contaminated areas or prevent extensive downslope dispersion during runoff;

- barriers such as earthen caps, surface seals, and walls to prevent extensive lateral dispersion of the contaminant or eliminate infiltration of water and leaching;
- land stabilization through the use of vegetation or well-compacted, fine-grained soil covers to reduce runoff velocities (and erosion) and minimize infiltration;
- monitoring of air, soil, surface water, and groundwater to detect its movement and dispersal in the environment, define the physical extent of contamination, and determine the effectiveness of decontamination procedures (when the problem no longer exists);
- security to prevent unauthorized entry into the site;
- treatment, including <u>in situ</u> biological, chemical, and physical treatment techniques, to degrade or detoxify contaminants or fix them in place;
- recordkeeping to characterize waste, evaluate monitoring data, record inspection results, and document maintenance performed, and;
- risk assessment to evaluate exposures, population-at-risk, health risks, and uncertainties associated with contaminant effects.

The costs of the above remedial actions will be quite high. The range of cleanup costs for various types of hazardous substances are approximately:

- \$2.20/kg (\$1/1b) of soluble substance on land,
- \$1.85 per liter (\$7/gal) of floating substance on water,
- \$53 to \$320 per liter (\$200 to \$1200/gal) of insoluble substance on land or sinking substance on water,
- \$160 per liter (\$600/gal) for PCB spills,
- \$860 to \$8200/ha (\$350 to \$3300/per acre) for fencing off land,
- \$1,200 to \$7,400/ha -(\$500 to \$3000 per acre) for land stabilization with vegetation,
- \$2,200 to \$12,000/ha (\$900 to \$5000 per acre) for deep plowing,
- \$5,900 to \$35,000/ha (\$2400 to \$14,000 per acre) for a soil cover or cap,

- \$4,000 to \$25,000/ha (\$1600 to \$10,000 per acre) for soil removal and onsite disposal, and
- \$270,000 to \$1,500,000/ha (\$110,000 to \$600,000 per acre) for soil removal and offsite disposal.

Historically, remedial actions for contaminated lands have largely been based on "being safe, rather than sorry" instead of on health risk assessments or on "acceptable" government standards. This is not to say that risk assessments or government standards have not been used in the planning process, but rather, that protection of the public (e.g., evacuation of the area), has been the overriding criterion. For example, in the cleanup of radioactive debris, soils, and vegetation in the Enewetok Atoll Islands after many years of nuclear weapons testing, the final criteria used by the U.S. Government were to expend greatest effort in areas of human occupancy, less in agricultural areas, and least in unmanaged areas.

In the case of a localized contamination as in the dioxin spill in Seveso, Italy, it may be prudent to identify areas of increasing exposure to the contaminant and base remedial action intensity accordingly. Of course, costs of cleanup and decontamination are important considerations, and decreased efficiency of cleanup with considerable increase in cost may set a practical limit to the effort. However, every land contamination occurrence will be unique and will require individual assessment. There will be no universal set of rules, but rather, a set of guidelines, such as put forth in this document, which may be followed in the development of a particular, individualized remedial action plan.

#### 5.2 RESEARCH NEEDS

Estimates of postnuclear attack external radiation doses have been made; however, integrated assessment of population doses from inhalation and ingestion pathways has not been done. It is not clear whether internal or external doses would predominate in the years following a nuclear war. Methods for performing such assessments are available, but assumptions concerning postattack population dynamics, agricultural practice, land decontamination, etc., are uncertain. Nevertheless, such an integrated assessment is recommended to determine the scope of the problem, quantify the relative contribution of internal exposure pathways (and therefore, the impact of remedial actions), and rank remedial action options.

Informed response to and management of land contaminated by a hazardous chemical is dependent on accurate information on the chemical's environmental behavior, persistence, and toxological properties. Monitoring and analytical support of remedial actions is possible only if certified standards, laboratory methods, and quality control procedures are available. For most hazardous chemicals such is not the case. It is recommended that efforts to compile existing information on solubility, octanol-water partitioning, soil-water distribution, and environmental persistence of hazardous chemicals continue. Simple laboratory studies on soil persistence, leachability, partitioning, etc., could supplement existing information. Finally, a program to establish a national repository of certified standards for important hazardous wastes should also be implemented. Along with the

standards, a peer reviewed set of quality control and analysis procedures should also be established.

Since it is recognized that it will be impossible to compile data on every hazardous chemical, it will be important to establish and verify relationships between a chemical's physical properties, e.g., molecular weight, functional groups, solubility, etc., and its environmental behavior. Such structure-activity relationships have been established, but large uncertainties are associated with them. An effort to reduce these uncertainties and improve predictive capabilities of present-day models is needed. Refinement of the models could be accomplished not only through the input of additional information, but also through multiple regression techniques, uncertainty analysis, and model validation. Such efforts would ultimately lead to the identification of additional information and research needs.

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#### 20. ASSTRACT (Continue on reverse edds if reseconty and identify by block number)

This report examines the various sources of radiological land contamination; its extent; its impacts on man, agriculture, and the environment; countermeasures for mitigating exposures; radiological standards; alternatives for achieving land decontamination and cleanup; and possible alternatives for utilizing the land. The major potential sources of extensive long-term land contamination with radio-nuclides, in order of decreasing extent, are nuclear war, detonation of a single nuclear weapon (e.g., a terrorist act), serious reactor accidents, and nonfission nuclear weapons accidents that disperse the nuclear fuels (termed "broken arrows"